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Bassetite and uranospathite.[1]

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[Read by Dr. A. F. Hallimond, March 25, 1954.]

Introduction.

BASSETITE and uranospathite were described as new uranium minerals from Cornwall in 1915 by A. F. Hallimond, to whose paper[2] reference should be made to supplement the descriptions given here. The position of both minerals as relatives of autunite was recognized by Hallimond. Unfortunately, chemical analyses could not be obtained and the composition of these minerals has remained uncertain. No further information has been published about uranospathite. In 1940 Meixner[3] reported that microchemical tests of an apparently authentic specimen of bassetite from Cornwall indicated that the mineral was a hydrated iron uranyl phosphate. Later George[4] briefly described under the name 'iron uranite' a more or less altered mineral from New Mexico that also appeared from qualitative tests to be an iron member of the autunite family. The colour and indices of refraction of this material were so different from those given by Hallimond for bassetite, however, that both it and the material examined by Meixner were considered to represent a different species. Recently, Branche, Chervet, and Guillemin[5] have referred a mineral from the Puy-de-Dôme district,

[1] Contribution from the Department of Mineralogy and Petrography, Harvard University, no. 346. Studies on uranium minerals, no. 17.

[2] A. F. Hallimond, Min. Mag., 1915, vol. 17, p. 221.

[3] H. Meixner, Chemie der Erde, 1940, vol. 12, p. 433. [M.A. 7–529.]

[4] d'A. George, Mineralogy of uranium and thorium. U.S. Atomic Energy Comm., 1949, RMO–563.

[5] G. Branche, J. Chervet, and C. Guillemin, Bull. Soc. Franç. Min. Crist., 1951, vol. 74, p. 457. [M.A. **11**–542.]

France, to bassetite on the basis of optical characters, although they note that qualitative tests showed the presence of only traces of iron.

Several years ago, Dr. Hallimond kindly forwarded to the writer authentic specimens of both bassetite and uranospathite. Examination of this material led to the recognition of additional specimens in the Harvard collection, and a fuller description of both minerals can now be given.

Bassetite.

Semi-quantitative spectrographic analyses[1] showed that type bassetite contained only Fe, U, and P as major constituents, together with traces of Mg, Cu, Al, and Si. X-ray powder patterns proved to be virtually identical with that of fully-hydrated saléeite, the Mg member of the torbernite group, and resembled, although less closely, the patterns of the other members of this group. The X-ray powder spacing data are given in table I. There is no similarity to the general type of pattern afforded by the lower hydrates of the metatorbernite group. Bassetite thus appears to be the ferrous iron member of the torbernite group, as was indicated by Meixner, but the original description, as well as optical and X-ray single-crystal study of the new crystals described below, indicates that the true symmetry is not tetragonal but monoclinic. Kahlerite, recently described by Meixner[2] from Hüttenberg, Carinthia, is chemically the arsenate analogue of bassetite. It is tetragonal in morphology, with biaxial optical anomalies.

Optically, the material received from Hallimond and five additional specimens later identified in the Harvard collection gave concordant measurements on air-dried samples, but with indices of refraction and extinction angles considerably higher than those originally reported by him (table II). The differences, however, are found to be due to variation in the content of zeolitic water in the mineral, a feature already well known in the minerals of the torbernite and metatorbernite groups.[3] When the samples were dried for two years at room-temperature over $CaCl_2$ the indices of refraction increased markedly. On the other hand,

[1] Kindly made by C. Annell and K. Valentine of the Trace Elements Laboratory, U.S. Geological Survey, Washington, D.C.

[2] H. Meixner, Der Karinthin, 1953, no. 23, p. 277; Chemie der Erde, 1940, vol. 12, p. 433.

[3] J. Beintema, Rec. Trav. Chim. Pays-Bas, 1938, vol. 57, p. 155 [M.A. **7**–237]; A. F. Hallimond, Min. Mag., 1916, vol. 17, p. 326, and 1920, vol. 19, p. 43; E. W. Nuffield and I. H. Milne, Amer. Min. 1953, vol. 38, p. 476 [M.A. **12**–281]; C. Frondel, Amer. Min., 1951, vol. 36, p. 671 [M.A. **11**–412].

the indices and the extinction angle decreased virtually to the values of Hallimond when the air-dried samples were kept for two years at room-temperature over H_2O. The samples kept over water became surficially turbid and brown due to oxidation of the ferrous iron. Hallimond's original material probably was near the fully hydrated condition, and

TABLE I. X-ray powder spacing data for bassetite.

(Copper radiation, nickel filter, in Ångströms.)

d.	*Int.*	*d.*	*Int.*	*d.*	*Int.*	*d.*	*Int.*
8·59	6	2·96	3	2·13	1	1·557	1
4·89	10	2·85	2	2·07	½	1·429	½
4·66	½	2·72	½	1·921	½	1·374	½
4·24	3	2·55	½	1·830	2	1·316	½
4·05	3	2·46	2	1·783	½	1·191	1
3·46	10	2·34	½	1·741	2	1·162	½
3·32	½	2·26	½	1·708	½	1·128	1
3·10	1	2·20	6	1·627	1	1·106	1

TABLE II. Optical data for bassetite.

(All measurements in Na-light.)

Sample.	α.	β.	γ.	Ext. angle.	2V.
Hallimond's 1915 data ...	—	1·574	1·580	−4°	large
Hallimond's new sample					
air-dried	—	1·606	1·610	+19	large
2 years over $CaCl_2$	—	1·627	1·665	—	large
2 years over H_2O	—	1·585	1·585	0	small
Harvard specimen, 101274					
air-dried	1·603	1·610	1·617	+18½	~ 90°
2 years over $CaCl_2$	—	1·632	1·67	—	large
2 years over H_2O	—	1·583	1·583	0	small
Harvard specimen, 88119					
air-dried	—	1·604	1·611	~ 20	large
Harvard specimen, 101276					
air-dried	—	1·603	1·607	~ 9	moderate
Harvard specimen, 101277					
air-dried	—	1·610	1·610	~ 8	small
George, 1949, New Mexico					
(dark brown)	1·60	1·65	1·66	?	40–50°

loss of water therefrom is accompanied by an increase in the extinction angle (a fact noted by Hallimond) and by increases in the indices of refraction, the partial birefringence $\gamma-\beta$, and 2V. The absorption of γ also increases relative to that of β. The optical differences between Hallimond's original material and the material later sent by him doubtless reflect the climatic differences between London and Cambridge, Mass. [See note appended to this paper.] The relatively high indices reported by George on what appears to be bassetite from New Mexico,

cited in table II, may be due to dehydration and, in part, to oxidation. The samples treated over $CaCl_2$ gave very poor X-ray patterns and their relation to the lower hydrates of the metatorbernite group could not be established; however, the phase change, if there is one, appears from the optical evidence to be reversible.

Bassetite from Cornwall is of two types. One consists of tabular crystals of pseudo-tetragonal appearance, with a yellow, brownish-yellow or, more commonly, bronze-yellow colour. The matrix usually is a black, rather porous gossan. The crystals vary from distinct single-crystals, of the sort measured by Hallimond, to aggregates of rectangular laths criss-crossing at right angles to give a lattice-like or chess-board appearance. The specimens described by Meixner are of the latter nature, and similar specimens are kept in various European and American collections. A specimen possibly of bassetite or autunite was figured in colour in 1806 by Sowerby.[1] Three specimens of the chess-board habit were found in the Harvard collection. One was unlabelled, another carried the notice 'Uranium Mineral? Cornwall. Talling, 1874', and the third specimen, although unlabelled, was entered in the Museum accession records as from Wheal Basset, Cornwall. Richard Talling (1820–83) of Lostwithiel was an English mineral collector and dealer, who supplied material to many collections. These specimens were identified as bassetite by optical, X-ray, and qualitative chemical study.

The second type of bassetite, comprising distinctly monoclinic single-crystals, was observed on only two specimens, both in the Harvard collection. These specimens are deeply oxidized pieces of pyritic vein matter, impregnated with clayey material of a greenish-grey colour, that contain small cavities and veinlets lined or filled with lamellar aggregates of yellow bassetite of the first type, together with tiny monoclinic crystals of an olive-green to olive-brown mineral. One of the specimens, no. 101274, carried a handwritten label 'Uranium? From Mr. Brooke. A. K. Barnett (Penzance) called on me a few weeks ago and said that he knew this mineral well as lime uranite. J. P. Jan. 31, 1886.' This specimen was acquired from the Calvert collection by Harvard in 1938, and it had earlier belonged, as appears from the label, to the English metallurgist and chemist John Percy (1817–89). The Mr. Brooke mentioned is the English crystallographer and mineralogist H. J. Brooke (1771–1857), from whose mineral collection, now preserved in Cambridge, England, came the bassetite twin crystal measured by Hallimond. The second specimen, no. 101275, carried a faded, handwritten label reading

[1] J. Sowerby, British mineralogy. London, 1806, vol. 2, p. 48, plate 126.

Yellow uranite. Huel Basset'. The two Harvard specimens correspond exactly, and there is little doubt that they were found in immediate association in Wheal Basset.

A chemical analysis by Mr. F. A. Gonyer, cited in table III, established the composition of the monoclinic olive-green crystals (and therefore of bassetite in general) as $Fe(UO_2)_2(PO_4)_2.nH_2O$. In the analysed sample n is about 8, and this value can be taken as representative of material kept in a warm, dry atmosphere. The small amount of Cu

TABLE III. Chemical analyses of bassetite and uranospathite.

	P_2O_5.	As_2O_5.	FeO.	CuO.	UO_3.	H_2O.	insol.	Total.
1.	15·26	—	7·73	—	61·51	15·50	—	100·00
2.	15·21	—	7·60	0·28	61·99	14·49	0·63	100·20
3.	4·50	14·59	—	7·57	54·47	18·87	—	100·00
4.	4·35	13·76	0·72	6·65	56·16	17·94	—	99·58

1. Theoretical weight percentages, $Fe(UO_2)_2(PO_4)_2.8H_2O$.

2. Bassetite, Wheal Basset, Cornwall. Olive-green monoclinic crystals. Harvard specimen 101274. F. A. Gonyer, analyst, 1949.

3. Theoretical weight percentages, $Cu(UO_2)_2\{(As,P)O_4\}_2.11H_2O$, with As:P = 2:1.

4. Uranospathite (?), Wheal Basset, Cornwall. Harvard specimen 98068. F. A. Gonyer, analyst, 1949.

reported doubtless is in substitution for Fe. A few crystals were observed that showed an indistinct colour zoning, with an accompanying variation in extinction angle from about 18° on the periphery to about 6° in the core, due to compositional variation of an unknown nature.

Morphological and X-ray single-crystal study indicates that bassetite is monoclinic in crystallization, although with very marked tetragonal pseudo-symmetry. The olive-green crystals are lozenge-shaped plates flattened on (010) and recall gypsum in habit (fig. 1). The interfacial angles could be measured only approximately due to the poor surface quality of the crystals. They are consistent within the limits of error with a tetragonal interpretation in which the plane of flattening is taken as (001) and alternate faces of forms other than (001) are undeveloped. The monoclinic axes $a\ b\ c$ then are equivalent to the tetragonal axes $a\ c\ a$ respectively. A list of the observed forms is given in both the monoclinic and tetragonal interpretations in table IV, and the relation of the two orientations is shown in fig. 1. The forms $a\ n\ q$ are small in size and often are lacking. Hallimond also concluded from morphological study of crystals of the first type that bassetite was monoclinic. His material had an almost square outline with large faces of the negative orthodome $(10\bar{1})$ and other forms not observed here. His elements, cited below, are

in fair agreement, but they differ from those for the present analysed crystals.

a.	*b*.	*c*.	*β*.	
0·3615	1	0·3662	90° 30′	(Hallimond, specimen 1)
0·3473	1	0·3456	90° 43′	(Hallimond, specimen 3)
0·409	1	0·411	90° 32′	(present study, X-ray)

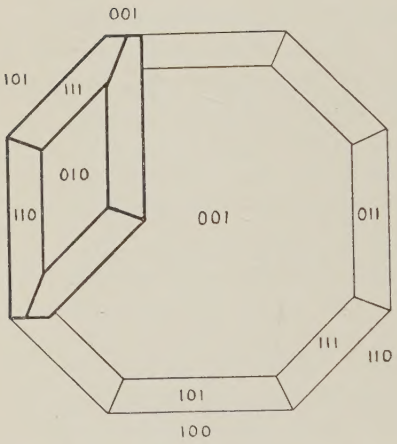

Fig. 1.

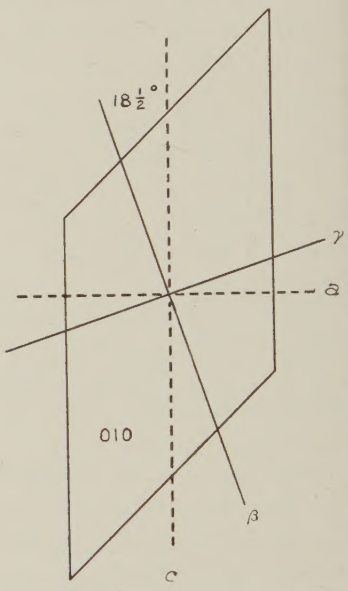

Fig. 2.

Fig. 1. Bassetite crystal projected on (010) (heavy lines), showing relation of the monoclinic and tetragonal interpretations.

Fig. 2. Optical orientation of bassetite. Olive-green crystals from Wheal Basset, Cornwall.

An X-ray single-crystal study by the Weissenberg method in both Cu and Fe radiation on crystals from the analyzed sample gave cell dimensions, cited below, consistent within the limits of error with tetragonal

a 6·98±0·04 Å.	*a*:*b*:*c* = 0·409:1:0·411
b 17·07±0·04	*β* = 90° 32′±5′ (Laue)
c 7·01±0·07	Cell contents $[Fe(UO_2)_2(PO_4)_2.8H_2O]$

symmetry. The cell dimensions in a tetragonal interpretation (*a* 7·0, *c* 17·07 Å.) approach those of the members of the torbernite group. The Weissenberg films were of poor quality, and the film symmetry was ambiguous due to slight smearing out of the spots which obscured intensity relations. All of the spots could be indexed as far as their

position was concerned in terms of a tetragonal cell. Rather sharp Laue photographs were obtained in W-radiation, with the beam travelling perpendicular to the plane of flattening, and these showed slight but definite departure from tetragonal symmetry in both the intensity and position of the innermost spots. Measurement of the axial directions of the Laue films established that β was 90° 32′ ± 5′.

TABLE IV. Angle table for bassetite.

(Angles calculated from the elements of the unit cell determined by X-ray measurement on olive-green crystals from Harvard specimen 101274.)

Monoclinic Interpretation.

$a:b:c = 0{\cdot}409:1:0{\cdot}411$; β 90° 32′; $p_0:q_0:r_0 = 1{\cdot}005:0{\cdot}411:1$;
$r_2:p_2:q_2 = 2{\cdot}433:2{\cdot}445:1$; μ 89° 28′; p_0' 1·005; q_0' 0·411; x_0' 0·009.

Forms.	ϕ.	ρ.	ϕ_2.	$\rho_2 = B$.	C.	A.
c 001	90° 00′	0° 32′	89° 28′	90° 00′	—	89° 28′
b 010	0 00	90 00	—	0 00	90° 00′	90 00
a 100	90 00	90 00	0 00	90 00	89 28	0 00
n 120	50 43	90 00	0 00	50 43	89 39½	39 17
m110	67 45½	90 00	0 00	67 45½	89 46	22 14½
d 101	90 00	45 24	44 36	90 00	44 52	44 36
p 111	67 56	47 34½	44 36	73 54	47 05	46 50
q 121	50 58	52 32½	44 36	60 00	52 08	51 55½

Tetragonal Interpretation.

$a:c = 1:2{\cdot}439$; $p_0:r_0 = 2{\cdot}439:1$.

Forms.	ϕ.	ρ.	A.	$\bar{M}$.
c 001	—	0° 00′	90° 00′	90° 00′
a 010	0° 00′	90 00	90 00	45 00
m 110	45 00	90 00	45 00	90 00
n 012	0 00	50 39	90 00	56 51½
d 011	0 00	67 42½	90 00	49 08
q 112	45 00	59 53½	52 17	90 00
p 111	45 00	73 50	47 13½	90 00

Monoclinic symmetry is also shown by the optical properties. The analysed crystals show parallel extinction when viewed along the plane of the (monoclinic) a and c axes and extinction angles of about +18° when viewed along the b axis. Their optical orientation is shown in fig. 2. The lamellar and platy bassetite of the first type also shows inclined extinction (table II). The sense of the extinction angle cannot be determined on crushed grains, since these are ordinarily bounded by virtually rectangular cleavage planes ($\beta \sim 90°$). The optic sign is negative.

The dispersion of the optic axial angle is strong, with $r > v$, and is not perceptibly inclined. Grains resting on (010) show anomalous interference colours in white light. The pleochroism is distinct, with α pale yellow to pale greenish-yellow and β and γ deep yellow to dark olive-brown or dark brown. The intensity of the pleochroism apparently varies somewhat with the degree of oxidation of the ferrous iron. The absorption of β and γ is about equal in relatively highly hydrated material, but both the absorption and the index of refraction of γ increase relative to β as water is lost. The lustre is bronzy inclining to metalloidal on (010) and is weakly vitreous on other faces. Bassetite has a perfect cleavage (010) and two distinct additional cleavages, that appear on crushed grains under the microscope as a system of cracks on (010) that parallel the a and the c axes. The absorption and cleavages conform in the tetragonal orientation to that observed in the members of the torbernite and metatorbernite groups. The hardness is $2\frac{1}{2}$. The specific gravity determined by the Berman balance on a coarse powder is 3·4 (3·6 calculated for $7\frac{1}{2}H_2O$). Hallimond gives the specific gravity of yellow crystals of the first type as 3·10. The agreement between these values is not satisfactory. Bassetite does not fluoresce in ultra-violet light.

The true symmetry of the members of the torbernite and metatorbernite groups has been under discussion for many years. Almost every member of these groups has been variously described as tetragonal, orthorhombic and pseudo-tetragonal, or monoclinic and pseudo-tetragonal. Lower than tetragonal symmetry has been ascribed chiefly on the basis of the optical properties, although attention also has been directed to slight departures in crystal angles from the requirements of tetragonal symmetry, to monosymmetric etching effects on the perfect cleavage, and to apparent differences in the ease of presumably equivalent cleavage planes. The X-ray structural study of Beintema (loc. cit.) on autunite showed that the basic structure of these minerals is tetragonal, and that the biaxial optical properties commonly observed are dependent on the number and symmetry of distribution of the zeolitic water molecules present. Later X-ray and dehydration studies on various members of these groups have confirmed this situation. In the present case there appears to be a definite distortion of the basic structure to give monoclinic symmetry, which is expressed in a distinctly monoclinic morphology, although there is no doubt but that the mineral is virtually isostructural with the torbernite group. This may also be the case with the ill-defined mineral trögerite, which appears to be a member of the

metatorbernite group although it is monoclinic in both crystal habit and optics.[1]

Uranospathite.

The following account of uranospathite is based on a type specimen from Redruth, Cornwall, supplied by Dr. Hallimond. The remarkably low indices of refraction that characterize the mineral were verified. The rapidity with which the mineral loses water unfortunately was underestimated, and the sample dehydrated irreversibly in lack of adequate precautions before all of the measurements on it were completed. Under the microscope, the fresh laths were biaxial negative with α perpendicular to the flattening and γ parallel to the elongation. There are distinct cleavages parallel and perpendicular to the elongation of the rectangular laths, and there is a perfect cleavage parallel to the flattening. The pleochroism is distinct with α colourless to pale yellow, β and γ deep yellow to greenish-yellow. The indices of refraction were rather variable, with the centres of the thicker crystals having the lower values. The measured values of α and γ are not quite as low as those of Hallimond, but this doubtless is due to partial dehydration. A sample exposed in the open air for a few hours increased considerably in indices, and when checked again overnight a larger increase was found with accompanying decrease in 2V. A sample kept for two years over $CaCl_2$ at room-temperature showed a further increase in indices, with 2V 0°, and the material was quite uniform in its optics. The decrease in 2V on dehydration also was noted by Hallimond. Another sample that had been air-dried for four weeks, and then had $\gamma \sim 1{\cdot}59$, was kept for two years over H_2O at room-temperature. This sample decreased in indices, but not to the original low values. The optical measurements are summarized in table V.

An X-ray powder pattern of the fresh material with low indices proved to be virtually identical with that of fully-hydrated synthetic zeunerite,[2] but with two additional lines at *d* 14·6 and *d* 7·56. Material that had been dehydrated by exposure to air or $CaCl_2$, on the other hand, was found to give the X-ray pattern of metazeunerite. A metazeunerite pattern also was afforded by material that had been air-dried and then kept over H_2O for two years, indicating that the phase change on

[1] A. Schrauf, Min. Mitth., 1872, p. 185; V. Goldschmidt, Zeits. Kryst. Min., 1899, vol. 31, p. 468.

[2] The synthetic material described by J. W. Frondel, Amer. Min., 1951, vol. 36, p. 249, was used for comparison. [M.A. **11**–323.]

dehydration is not reversible. The same behaviour has been found in metazeunerite itself. A semi-quantitative spectrographic analysis of the sample showed it to be an arsenate-phosphate of copper and uranium, with As > P.

With the aid of the above information, a specimen in the Harvard collection labelled 'Autunite? Wheal Basset' was found to be probably identical with dehydrated uranospathite. This specimen, originally from the Calvert collection earlier mentioned, showed fan-like groups of

TABLE V. Optical data on uranospathite.

Sample.	α.	β.	γ.	2V.
Hallimond's original data, 1915	[$\sim$ 1·49]	1·510	1·521	69°
Hallimond's new sample, freshly opened vial	1·485–1·495	1·522–1·527	1·527–1·532	55–60°
after 4 weeks in open air	1·558	1·591–1·593	1·593–1·595	20–30
after 2 years over $CaCl_2$	—	1·612	1·612	0
after 2 years over H_2O	—	1·54–1·57	1·54–1·57	0–10
Harvard specimen, air-dried	1·56	1·594–1·604	1·594–1·604	0–5
after 2 years over $CaCl_2$	—	1·614	1·614	0
after 2 years over H_2O	—	1·565–1·575	1·565–1·575	0–5

rectangular laths on a black porous gossan. The indices of refraction of the air-dried Museum specimen were relatively high, and corresponded not with the values of fresh uranospathite but with the values obtained after the latter mineral had been air-dried for four weeks. The refractive indices of material held for two years over H_2O and over $CaCl_2$ also were virtually identical with those of Hallimond's material when similarly treated (table V). A chemical analysis by Mr. F. A. Gonyer of the Harvard specimen, cited in table III, established the formula as $Cu(UO_2)_2\{(As,P)O_4\}_2.11H_2O$, with As:P = 2:1. This composition accords with the semi-quantitative spectrographic analysis made on the type uranospathite. The Harvard specimen gave the X-ray pattern of metazeunerite, and the analysis establishes it as an arsenic-rich member of a series between metazeunerite and metatorbernite.

The dehydration product of Hallimond's uranospathite thus appears to be identical with metazeunerite. This view might imply that the fully-hydrated material could be identical with zeunerite. The indices of refraction found for the fully-hydrated material, however, are considerably lower than those reported for zeunerite or torbernite. Unless zeunerite and torbernite may contain much more zeolitic water in the fully-hydrated state than has been recognized, uranospathite may represent a still higher hydrate in this group of minerals. The writer also has

been informed by Dr. C. Guillemin of the Laboratory of Mineralogy of the Sorbonne that a specimen of uranospathite supplied to him by Dr. Hallimond was found by qualitative chemical tests not to contain copper. It seems desirable to retain the name uranospathite in a provisional status until these matters are clarified.

Note by A. F. Hallimond (February 1954).

I am grateful to Professor Frondel for the opportunity of commenting on this paper. It is indeed satisfactory to have the present measurements and analyses on carefully studied material.

Bassetite. The author remarks on the substantial differences in density and axial ratios between my original measurements and those on his partially dehydrated material. I do not think they are accidental.

The values may be compared as follows (tetragonal representation).

	c:*a*	Sp. gr.		*c*:*a*	Sp. gr.
Torbernite (H.)	2·974	3·219	Metatorbernite (H.)	2·28	3·67
Bassetite (H.)	2·89	3·10	Bassetite, Harvard (F.)	2·44	3·4

My original bassetite is isomorphous with torbernite (both fully hydrated). The new bassetite (with only $8H_2O$) approaches the values for metatorbernite. I should regard this as due to morphotropic contraction of the *c*-axis on dehydration, analogous with that which I proved for torbernite (1920). This may well be the case even if the present altered mineral has not assumed complete 'meta-' structure.

Uranospathite. I would agree that the density and optical data exclude the possibility of this mineral being either zeunerite or metazeunerite. It is almost certainly a higher hydrate than $12H_2O$. The yellow colour of my original mineral is unlike that of a copper compound. It seems quite possible that uranospathite is one of a series of higher hydrates of the same uranyl compounds as are present in bassetite, &c.

Dehydration. Work now in progress on these and other uranium minerals in the Museum of Practical Geology (South Kensington), by S. H. U. Bowie and R. Harrison, indicates that considerable alteration has taken place in some species. This is no doubt connected with the transfer (about 1933) of the collection from the old Jermyn Street building. It was there stored in a basement where the atmospheric conditions were not so far removed from those of their original situation in the mine, while the present storage is in a top floor of the Museum, which is centrally heated.

The orthoclase-microcline inversion.

By Wm. Scott MacKenzie, M.A., B.Sc., Ph.D.

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[Taken as read January, 1954.]

Introduction.

THE association of orthoclase[1] and microcline in the same rock has been commented on by many writers, and from these occurrences attempts have been made to deduce the relative stability of these two low-temperature forms of potash-felspar. Almost all investigators have concluded that microcline is the lower-temperature modification, but difficulties in explaining the relationship of adularia to orthoclase and microcline have been encountered.

The identification of the monoclinic potash-felspar in association with microcline is almost always based on optical properties, viz. straight extinction in the zone [010] or the absence of visible multiple twinning. These are necessary conditions, but are not in themselves sufficient evidence of a monoclinic felspar lattice. X-ray powder diffraction patterns are sometimes effective (MacKenzie, 1952), but detection of very slight departures from monoclinic symmetry may require single-crystal X-ray methods.

In discussing the lattice and twinning of microcline, Laves (1950) states that the relationship between the pericline and albite twinning can be explained only on the assumption that the microcline crystallized with monoclinic symmetry and subsequently acquired triclinic symmetry. Many petrologists may find difficulty in accepting this hypothesis from their experience of the occurrence of microclines, but the evidence given by Laves from the material which he investigated is very good. In one of the specimens to be described here, orthoclase is actually present along with microcline, and the relationship between pericline and albite twinning in the microcline appears to be identical with that described by Laves (1950). There seems no firm reason, however, to extend the hypothesis to untwinned microclines.

[1] The term orthoclase is used throughout this paper to indicate low-temperature monoclinic potash-felspar.

Variations in the microcline lattice.

In 1950 Laves considered that microcline and triclinic adularia should be regarded as separate modifications of potash-rich felspar, and he gives lattice angles 'characteristic' of each form even in a more recent paper (1952), in which he suggests that there may be a continuous gradation from a truly monoclinic lattice to a triclinic lattice with angles α 90° 39′ and γ 87° 47′. These values are, however, very common in microclines. They represent the largest departure from monoclinic symmetry so far found, and following a suggestion by Dr. Laves (personal communication), the term 'maximum' microcline is used here to denote a potash-felspar having approximately these lattice angles.

Fig. 1 shows the X-ray spectrometer patterns of five samples of low-temperature, potash-rich felspar. Pattern A is of a truly monoclinic felspar, and the remaining patterns are produced by triclinic felspars. The peaks joined by broken lines represent reflections from the (130) and ($1\bar{3}0$) planes between 2θ values of 23° and 25° (Cu-$K\alpha$), and from the (131) and ($1\bar{3}1$) planes between 2θ values of 29° and 31°. The patterns are arranged in order of increasing departure from monoclinic symmetry.

The lattice parameters of specimens C, D, and E, measured from these patterns by the method of Donnay and Donnay (1952), are set down in table I. The relation between the separation of the 130 and $1\bar{3}0$ peaks and the angle γ^* is given by the following expression:

$$Q_{1\bar{3}0} - Q_{130} = 12a^*b^* \cos\gamma^*, \text{ where } Q_{hkl} = \frac{1}{d^2_{hkl}}.$$

The value of $2\theta_{130} - 2\theta_{130}$ is thus directly dependent on variations in γ^*, since variations in a^* and b^* are very small. The angles α and γ calculated from specimen E correspond fairly closely to the angles of a 'maximum' microcline. The value of $2\theta_{1\bar{3}0} - 2\theta_{130}$ (Cu-$K\alpha$) for this mineral is 0·84°.

The compositions of these five minerals, calculated from the chemical analyses, are given in fig. 1. In samples B, D, and E the peak at about $2\theta = 28°$ (Cu-$K\alpha$) indicates the presence of albite as a separate phase. This is the strongest peak in the powder diffraction pattern of low-temperature albite[1] and is made up of reflections from 040, 002, and 220. Thus the compositions do not represent the potash phase alone and cannot be used in a study of the microcline lattice without correcting for the amount of albite present as a distinct phase. If a large amount of low-temperature albite is present, the 111 reflection of the albite may

[1] From this peak alone it is impossible to distinguish between high- and low-temperature albite.

obscure the 130 reflection of orthoclase, but in these patterns this difficulty does not arise.

The lattice parameters of sample B have not been determined, because

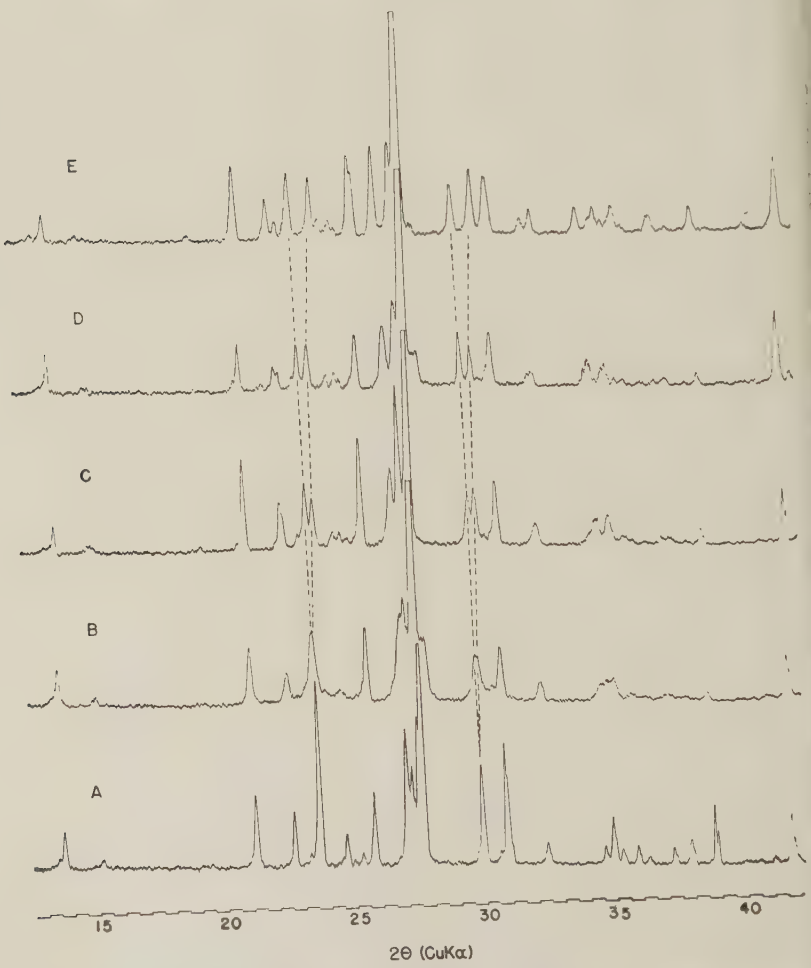

Fig. 1. Powder X-ray diffraction patterns of low-temperature alkali-felspars. Pattern A is of a monoclinic felspar and patterns B, C, D, and E are of microclines with varying departure from monoclinic symmetry. The peak at about 28° (Cu-$K\alpha$) in patterns B, D, and E is due to the presence of albite as a separate phase.

The compositions of these minerals from chemical analyses are: (A–D, Spencer, 1937) A, $Or_{84\cdot5}Ab_{13\cdot8}An_{1\cdot7}$; B, $Or_{81\cdot5}Ab_{18\cdot1}An_{0\cdot4}$; C, $Or_{85\cdot9}Ab_{12\cdot7}An_{1\cdot4}$; D, $Or_{83\cdot3}Ab_{14\cdot9}An_{1\cdot8}$; E (Blue Mt. microcline, table II), $Or_{90\cdot5}Ab_{9\cdot2}An_{0\cdot3}$. (See p. 366, *Note*.)

the reflections which show the triclinic character of the lattice are not clearly resolved from one another. It is probable that the lattice angles of this mineral are close to those given by Laves (1952) as characteristic

TABLE I. Cell parameters of microclines.

	Specimen C.	Specimen D.	Specimen E.
a	$8{\cdot}56_6$	$8{\cdot}55_9$	$8{\cdot}57_4$
b	$12{\cdot}96_4$	$12{\cdot}97_6$	$12{\cdot}98_1$
c	$7{\cdot}21_1$	$7{\cdot}20_2$	$7{\cdot}22_2$
α	90° 17′	90° 23′	90° 41′
β	115 58	115 54	115 59
γ	89 11	88 47	87 30
α^*	90 05	90 09	90 18
γ^*	90 46	91 09	92 22

The revised orientation suggested by Laves (1951) has been adopted. Dr. S. W. Bailey, of the University of Wisconsin, has determined cell parameters of a specimen of the same material as sample C by single-crystal X-ray methods. These are given for comparison with the above values (Bailey, personal communication): a 8·5784, b 12·9600, c 7·2112, α 90° 18′, β 116° 02′, γ 89° 08′, α^* 90° 05′, β^* 63° 58′, γ^* 90° 50′.

TABLE II. Analyses of 'maximum' microclines. (Analyst, J. H. Scoon.)

	1.	2.	3.
SiO_2	64·46	64·58	64·40
Al_2O_3	18·55	18·68	19·09
Fe_2O_3	0·14	0·12	0·31
MgO	nil	nil	0·04
CaO	0·17	0·20	0·05
Na_2O	0·49	0·63	1·09
K_2O	16·07	15·68	15·26
H_2O-	0·06	0·07	nil
MnO	nil	nil	0·01
	99·94	99·96	100·25
Or (wt. %)	95·0	93·6	90·5
Ab	4·2	5·4	9·2
An	0·8	1·0	0·3

1. Microcline from a pegmatite in grennaite, Norra Kärr, Sweden. $Or_{95\cdot0}Ab_{4\cdot2}An_{0\cdot8}$.

2. Amazonstone from Pikes Peak, Colorado. The analysed sample was estimated optically to contain 2 % of albite as a separate phase; thus the composition of the potash phase is $Or_{95\cdot6}Ab_{3\cdot4}An_{1\cdot0}$.

3. Microcline from the nepheline-syenite of Blue Mountain, Ontario. The analysed sample was estimated optically and by X-rays to contain 2 % of albite. The corrected composition of the potash phase is $Or_{92\cdot5}Ab_{7\cdot2}An_{0\cdot3}$.

of triclinic adularia, but few petrologists, on seeing the uniform cross-hatch twinning covering the entire area of the crystals, would call this mineral adularia.

Specimens A, B, C, and D are from Dr. E. Spencer's collection of orthoclase- and microcline-microperthites (Spencer, 1937), and the

writer is indebted to Drs. N. L. Bowen and O. F. Tuttle for permission to use them. Specimen E is from the nepheline-syenite of Blue Mountain, Ontario, and its composition was calculated from a chemical analysis by Mr. J. H. Scoon (table II).

Felspar from the Bearpaw Mountains, Montana.

The felspars from the nepheline-syenite pegmatites of Rocky Boy stock in the Bearpaw Mountains, Montana, have been described by Pecora (1942). He suggested that the felspars in many of the pegmatites had been hydrothermally altered and noted that, in some cases, grains which appear homogeneous in plane polarized light have a 'splotchy' texture under crossed nicols. Professor C. E. Tilley noted a considerable variation in the size of the optic axial angle of what appeared, in plane polarized light, to be a homogeneous felspar crystal from one of these pegmatites. The specimen described here is not identical with any of the materials described by Pecora, but is of the same general type and under crossed nicols it shows the 'splotchy' texture mentioned by him.

Fig. 2 shows a photomicrograph of part of one of these felspar crystals under crossed nicols; a suggestion of multiple twinning can be seen in this photograph. The small included crystals are aegirine. The section is approximately normal to the acute bisectrix. The optic axial angle of the dark areas is moderate (30–40°); in the lighter areas it is much larger (70–75°). The optic plane is common to the whole crystal, but the position of the acute bisectrix differs with the size of the optic axial angle. It was not possible to make optical determinations of the symmetry of the various parts of the crystal, since the cleavages are not well enough defined in thin section to obtain measurements more accurate than $\pm 1°$. The (001) and (010) cleavages appear to be continuous throughout the whole crystal, thus emphasizing its apparent optical homogeneity in plane polarized light. An X-ray spectrometer powder pattern of this mineral shows that it is rich in potash and consists of microcline and orthoclase with no soda-felspar phase. Fig. 5*a* (p. 362) shows part of the X-ray spectrometer pattern.

Zero layer-line *a*- and *c*-axis Weissenberg photographs were made of crystal fragments of both the material of moderate optic axial angle and that of larger optic axial angle. The patterns of the former show only one set of reflections and are consistent with monoclinic symmetry. The pattern obtained from the fragment of larger optic axial angle (fig. 3) shows groups of three and five reflections instead of single reflections. The centre spot in every case corresponds with the appropriate spot on

the photographs of the monoclinic crystal, while the additional spots represent four triclinic components.

The stereogram in fig. 4 shows the relations between these five units as deduced from the X-ray photographs by Dr. J. V. Smith. The axes of the monoclinic component are denoted by the subscript M and those of

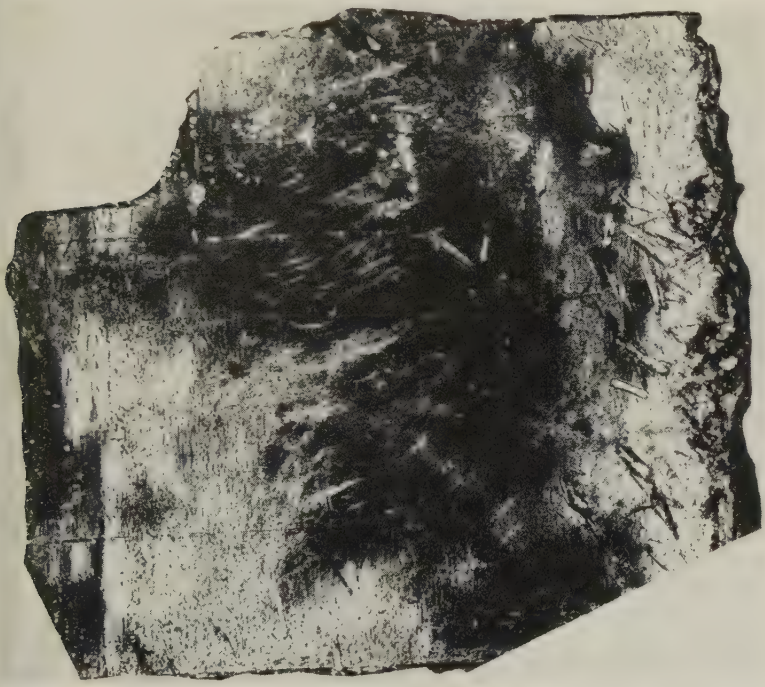

FIG. 2. Photomicrograph of a felspar from a nepheline-syenite pegmatite of Rocky Boy stock, Bearpaw Mts., Montana. The crystal is cut approximately normal to the a crystallographic axis. The dark areas are monoclinic and the lighter areas are triclinic. Crossed nicols: $\times 96$.

the triclinic components by the subscripts 1, 2, 3, and 4. The c^*-axis appears to be common to both monoclinic and triclinic parts. Equally inclined to the a^*-axis of the monoclinic component are two additional a^*-axes which lie in the a^*b^* plane of the monoclinic material. Two additional b^*-axes are equally inclined to the b^*-axis of the monoclinic material, and lie in the plane at 90° to the common c^*-axis. There are thus five separate orientations, of which one is monoclinic and four are triclinic with a common c^*-axis. Study of the stereogram reveals that it

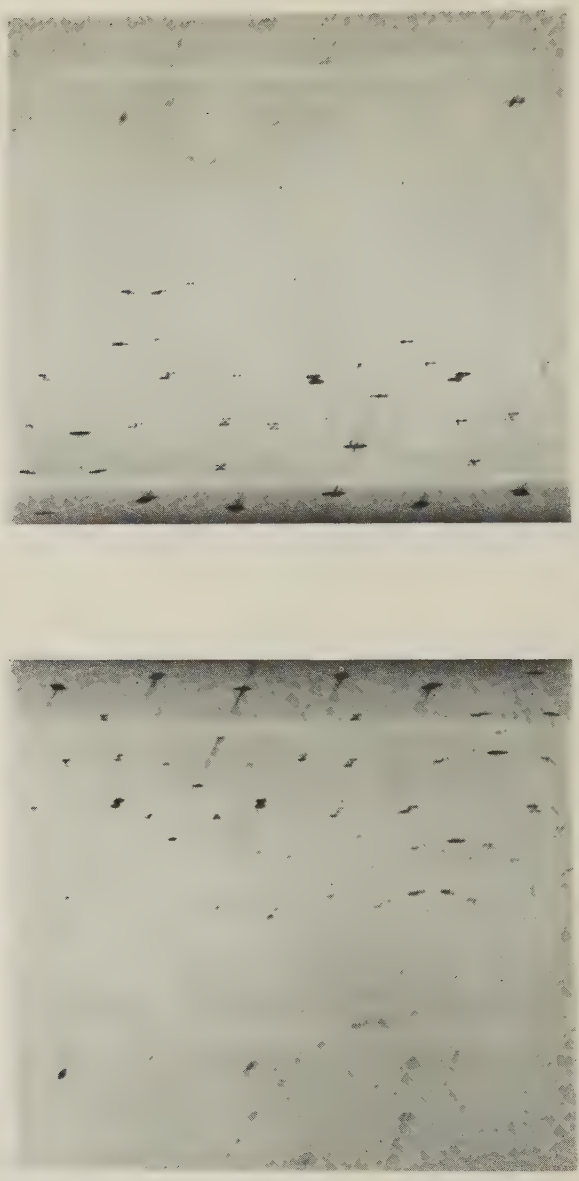

FIG. 3. Zero layer-line Weissenberg photograph taken about the c-axis of a fragment of the felspar from the Bearpaw Mts., Montana, using Cu-$K\alpha$ radiation. The reflections from the lattice planes of the five differently oriented units in this crystal are clearly shown and form small crosses in the photograph.

must be only an approximation, as two of the triclinic components evidently have b^* in common with the monoclinic material and therefore cannot have a common c^*-axis with it unless the 'triclinic' α^* is exactly 90°. The maximum departure from 90° of the angle α^* in a microcline, about 25′ according to Laves (1950), cannot be reliably detected in these Weissenberg photographs.

Laves (1950) has described the relationship between the separate

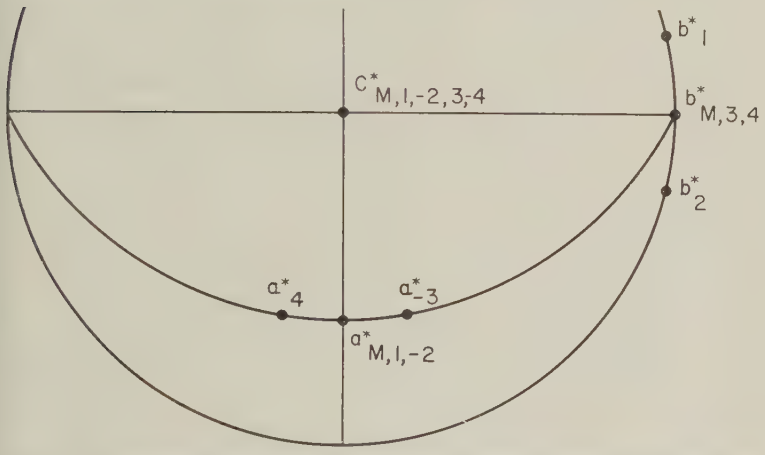

FIG. 4. Stereogram showing the approximate relation between the axes of the reciprocal lattices of the five different units of the felspar from the Bearpaw Mts., Montana. The subscript *M* refers to the monoclinic part and the subscripts 1, 2, 3, and 4 refer to the four triclinic parts. The angular relations in the stereogram are very much exaggerated for clarity.

triclinic units of a twinned microcline as due to albite and pericline twinning, the albite-twinned parts retaining the plane of symmetry of the original monoclinic material and the pericline-twinned parts retaining the axis of symmetry. From the stereogram in fig. 4 it can be seen that the triclinic parts 1 and 2 are in pericline twin relation and parts 3 and 4 are in albite twin relation. There is no evidence to suggest that the twinning arrangement differs from that proposed by Laves (1950), with the exception that the original monoclinic material is still preserved, and the close relationship between the orientations of the triclinic material and the monoclinic parent material can be seen in the X-ray photographs.

The lattice angles α and γ of this triclinic material, measured from the X-ray spectrometer pattern, do not show as large a departure from 90°

as in 'maximum' microcline, the value of $2\theta_{1\bar{3}0}-2\theta_{130}$ being 0·71° (Cu-$K\alpha$).

Felspar from paragneiss, Glen Urquhart, Scotland

This felspar occurs in an area of paragneisses which, according to Francis (1952), have been subjected to potash metasomatism. Almost all the grains show very uneven extinction, parts of individual crystals show cross-hatch twinning typical of microcline, and other parts are untwinned. This is quite a common feature of microcline, and, on the basis of extinction-angle measurements, many authors have attributed this to the association of orthoclase and microcline. A small amount of microperthitic albite is irregularly distributed in many of the crystals.

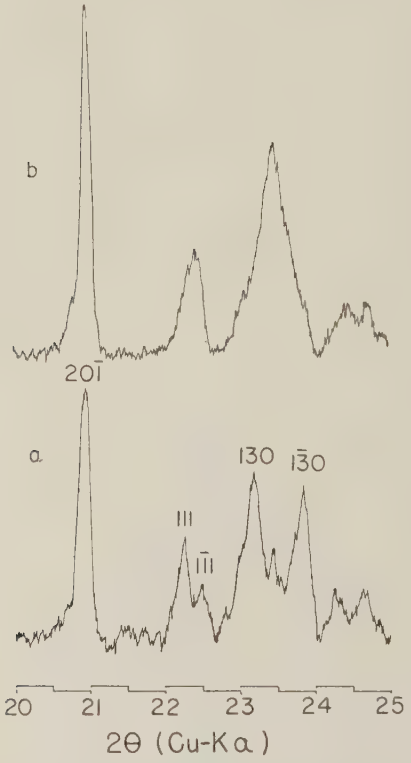

Fig. 5. Parts of the X-ray spectrometer patterns of the felspars from (*a*) the Bearpaw Mts., Montana, and (*b*) Glen Urquhart, Scotland. In pattern (*a*) the presence of orthoclase is indicated by the reflection between the 130 and 1$\bar{3}$0 reflections of microcline. In pattern (*b*) the variable lattice of microcline is shown by the broad peaks representing the 130 and 1$\bar{3}$0 reflections and the 111 and 1$\bar{1}$1 reflections. By contrast the 20$\bar{1}$ peak is sharp and well defined.

Laves (1950, p. 553) has discussed this question of uneven extinction and optically monoclinic areas in a microcline from Madagascar, and has presented two possible explanations; viz. optical deviation from monoclinic symmetry is a function of either (*a*) the lattice angles α and γ, or (*b*) the ratio of left-hand to right-hand albite twin positions if the material is submicroscopically twinned. Since he found differences in the intensities of the X-ray reflections from the two parts of the twin, he adopted the

latter explanation as the correct one for the Madagascar microcline.

Part of an X-ray spectrometer pattern of the Glen Urquhart felspar is reproduced in fig. 5*b*. The single broad peak shown by this mineral for the 130 and $1\bar{3}0$ reflections indicates that, although monoclinic or very nearly monoclinic material is predominant, microcline is present and there is considerable variation in its lattice. In this particular case, then, the optical deviation from monoclinic symmetry may well be a measure of the structural deviation. Although it seems quite likely, it cannot be established from this pattern that a truly monoclinic felspar is present, since a very slight departure from monoclinic symmetry will not cause sufficient separation between the $1\bar{3}0$ and 130 peaks in the powder pattern for these to be clearly resolved from each other.

Discussion.

From these two examples it would appear that there are two different ways in which triclinic and monoclinic potash-felspar may coexist: (*a*) the monoclinic material is associated with triclinic material having a fixed and fairly large departure from monoclinic symmetry, or (*b*) the monoclinic material is associated with triclinic material which shows all gradations in its deviation from monoclinic symmetry.

Barth (1934) first suggested that the differences between orthoclase and microcline are the result of disordering and ordering of the Si and Al atoms, microcline being the form in which ordering is more nearly complete. Laves (1950, 1952) elaborated on Barth's hypothesis and suggested that below 700° C. the values of the axial angles α and γ in potash-felspar increasingly deviate from 90°, the deviation being a function of the degree of ordering of the Si and Al atoms. Felspars showing intermediate structural states between a monoclinic lattice and that of a 'maximum' microcline might be described by Laves as adularia (1952, p. 445). In an attempt to explain the relationship of adularia to orthoclase and microcline, he suggested that, although adularia is generally considered to have been formed at very low temperatures, it might be a partially disordered metastable form resulting from the rate of growth being sufficiently high 'that the ordering forces are overwhelmed during crystallization'. Laves himself states, however, that 'no natural albites have been reported that display a behaviour that could be considered to be due to an intermediate state between albite and analbite'[1] (1952,

[1] Although petrologists may not have reported intermediate states between high- and low-temperature albite, these probably do exist in volcanic and hypabyssal

p. 446). Albites from low-temperature environments characteristically show very sharp extinction. The presence of any high-temperature albite would be easy to detect optically because of the considerable differences in the extinction angles and optic angles of the two forms.

Although Laves (1952) has inserted a footnote to the effect that the cell angles which he gives for microcline may be somewhat variable depending on the soda content, none of the discussions of the relationship between monoclinic and triclinic potash-felspar appear to have taken full account of the possibility that the symmetry may be controlled by the chemical composition. This omission can probably be explained by the difficulty of obtaining accurately the composition of the potash phase in microperthites.[1] It seems possible that variations in the lattice of microclines might be directly related to their chemical composition, as has been shown to be the case in the anorthoclase series (Donnay and Donnay, 1952). Since microclines are almost always microperthitic, it is difficult to find material from which the soda phase can be completely separated and the composition of the potash phase determined chemically. A solution to this problem might be found in an estimate, from the X-ray spectrometer pattern, of the amount of albite present as a separate phase in each analysed specimen.

Mixtures containing 10, 5, 4, and 2 % of albite were prepared from natural samples of a rather pure homogeneous orthoclase and a low-temperature albite, previously sieved to the same grain-size. X-ray spectrometer patterns of these mixtures showed that the height of the peak representing the 002, 040, and 200 reflections of albite is a function of the concentration of albite. This peak was easily detected in the mixture containing only 2 % of albite. Because of the uncertainty regarding the minimum size of the units of albite in microcline-perthites, the accuracy of this method for determining the amount of albite in a perthite has not been investigated in detail.

In three new analyses of 'maximum' microclines from which most of the albite was separated (table II), the amount of soda-felspar in solid solution does not exceed 7·5 % after correcting for albite visible under the microscope. Felspars like those described by Laves (1950) as triclinic adularia having only a small departure from monoclinic symmetry

rocks. Optical inhomogeneity generally would be attributed to chemical zoning, which, of course, may also be present.

[1] Vogt (1926), Andersen (1928), and Spencer (1937) have remarked on the limited range of composition of microcline-microperthites, but Spencer states that more extreme compositions could possibly have been found in material which was discarded as not suitable for his accurate optical investigations.

are frequently non-perthitic. In the literature, chemical analyses of adularias show that these have between 10 and 20 % of soda-felspar in solid solution. Specimen C (fig. 1) probably has less than 1 % of albite as a separate phase, so it will have 13 % of soda-felspar in solid solution.

Returning to a consideration of the two specimens described in this communication, it appears that the felspar from the Bearpaw Mountains has changed from a monoclinic lattice directly to a triclinic lattice of fixed lattice parameters with very little material of intermediate structural states. There is no evidence of a soda phase either under the microscope or in the X-ray diffraction patterns. According to the argument proposed here, this type of change is possible only because the original monoclinic felspar has a very low soda content. The Glen Urquhart felspar, on the other hand, shows gradational lattice parameters, and the presence of small blebs of albite indicates that some exsolution has taken place. It is conceivable that these two factors are related and that only by exsolution of albite to reduce the amount held in the potash phase can the lattice angles approach those of a 'maximum' microcline.

One piece of evidence which may conflict with the hypothesis of chemical control of the lattice of microclines was given to me recently by Dr. O. F. Tuttle (personal communication). An X-ray spectrometer pattern was made of a microcline-microperthite and the distance between the (130) and ($1\bar{3}0$) peaks was measured on the chart; the presence of albite lines in the pattern was noted. The specimen was heated for different periods of time until albite lines could no longer be detected in the X-ray pattern, and it was found that the separation of the (130) and ($1\bar{3}0$) peaks had not altered within the limits of error in measurement. The albite had been taken into solid solution in the potash phase. Dr. Tuttle has remarked, however, that the homogeneous phase produced may be metastable, so this cannot be considered as evidence against the hypothesis proposed above.

The writer believes that both the character of the lattice and the very existence of the lattice of microcline are controlled chiefly by the chemical composition of the potash phase, which, in most cases, is controlled by the extent to which the exsolution of soda-felspar has proceeded. If metamorphism is capable of promoting exsolution, and many petrologists have expressed the view that it is, a possible reason for the occurrence of microcline as the stable form of potash-felspar in rocks which have been subjected to regional metamorphism is that the mineral is almost pure $KAlSi_3O_8$.

The hypothesis of chemical control of the lattice angles of microclines

may not be completely at variance with the idea that the change from monoclinic to triclinic symmetry is connected with the ordering of the Si and Al atoms. Perhaps the Si and Al atoms cannot order beyond a certain limit until the soda and potash phases have unmixed to the extent that each is almost a pure end-member of the solid solution series. This limit of the ordering of Si and Al may be represented by orthoclase occurring as a discrete phase or by the monoclinic potash phase in orthoclase-microperthites.

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Note.—Fig. 1, B and D: lattice parameters and amount of exsolved albite are variable in different fragments. A, B, C, D are Spencer's D, W, U, E; his analyses are recalculated to 100%.

Zoned plagioclases in layered gabbros of the Skaergaard intrusion, east Greenland.

By J. M. Carr, M.A., D.Phil., F.G.S.

[Taken as read March 25, 1954.]

I. Introduction.

A LIMITED amount of careful optical work has been carried out on three analysed felspars of gabbros belonging to the main layered series of the Skaergaard intrusion, east Greenland (Wager and Deer, 1939). The work was begun with the intention of providing precise optical data to be used in further defining the relationship between composition and optics. The primary precipitate felspar crystals, hitherto thought to be devoid of zoning, were found, however, to possess a zoning which prevents the data being used in this way. Nevertheless, the zoning itself proved of interest. The majority of crystals possess a mild degree of normal zoning, but in one of the three specimens a small proportion of the primary precipitate felspars are oscillatory-zoned. The latter feature seems of particular significance.

II. Description of the primary precipitate zoning.

Wager and Deer showed that the layered gabbros are composed of primary precipitate crystals in a matrix of interprecipitate material. The primary precipitate crystals formed as successive crops of crystals from the well-stirred magma, while the interprecipitate material crystallized with falling temperature from the trapped interstitial liquid. The interprecipitate felspar is estimated to form 20 % or less of the total plagioclase. In thin section a strong normal zoning[1] at once distinguishes it from the euhedral primary precipitate crystals. The latter are enclosed, either partially or completely, within interprecipitate felspar. The interprecipitate zoning, often culminating in highly sodic felspar, is a general feature of the layered gabbros and requires no further description or explanation.

The primary precipitate zoning of the three felspars which have been chemically analysed (Wager and Mitchell, 1951, table 5) has been determined by methods described in the appendix, and the results are summarized in table I, together with the calculated normative composition

[1] Throughout this account the term 'normal zoning' is used in the accepted sense of an outward trend to a more sodic composition.

TABLE I. The range of primary precipitate zoning in the investigated plagioclase felspars.*

Specimen.	Normative composition by analysis†		(001) α′	(001) γ′	(010) α′	(010) γ′	Range of An % determined by R.I. method.	Range of An % determined by Zone method§	Probable zonal range.‖	Type of zoning.
4145 Ferrohortonolite-ferrogabbro height 2200 m.	$An_{38}Ab_{60}Or_2$	Range of R.I.‡	1·5498–1·5480	1·5551–1·5538	1·5493–1·5469	1·5545–1·5515	43–39	—	$An_{43} \rightarrow An_{37}$	Normal
		No. of flakes used	24	20	25	58				
		Normative % An	43 40	42–39	43–39	44–39				
2580 Hortonolite-ferrogabbro height 1800 m.	$An_{41}Ab_{56}Or_3$	Range of R.I.	1·5520–1·5490	1·5565–1·5553	1·5510–1·5492	1·5553–1·5531	46–42	46–40	$An_{46} \rightarrow An_{40}$	Either normal or oscillatory-normal
		No. of flakes used	18	11	20	8				
		Normative % An	46 42	44 42	45 42	46 42				
4077 Hypersthene-olivine-gabbro height 500 m.	$An_{55}Ab_{43}Or_2$	Range of R.I.	1·5590–1·5573	1·5654–1·5632	1·5685–1·5568	1·5640–1·5615	59–55	60–52	$An_{59} \rightarrow An_{52}$	Normal
		No. of flakes used	16	18	11	15				
		Normative % An	58–55	59–55	58–55	60–56				

* All values for An are in weight %.

† Including interprecipitate felspar. Analyst, W. A. Deer (Wager and Mitchell, 1951, p. 146).

‡ Refractive indices ±0·001.

§ The range for specimen 2580 concerns an oscillatory-zoned crystal.

‖ For the calcic limit of each range the estimated degree of error is ±1 part An, whilst the sodic limits are ±2 parts An except in the case of 4145, for which the sodic value is only approximate.

representing the bulk felspar composition of the rock. From these data it is concluded that the zoning of the primary precipitate crystals involves a change in composition of no more than six or seven parts anorthite.

Of the three gabbros from which felspars have been taken, the lowest in the main layered series is the hypersthene-olivine-gabbro 4077. Like

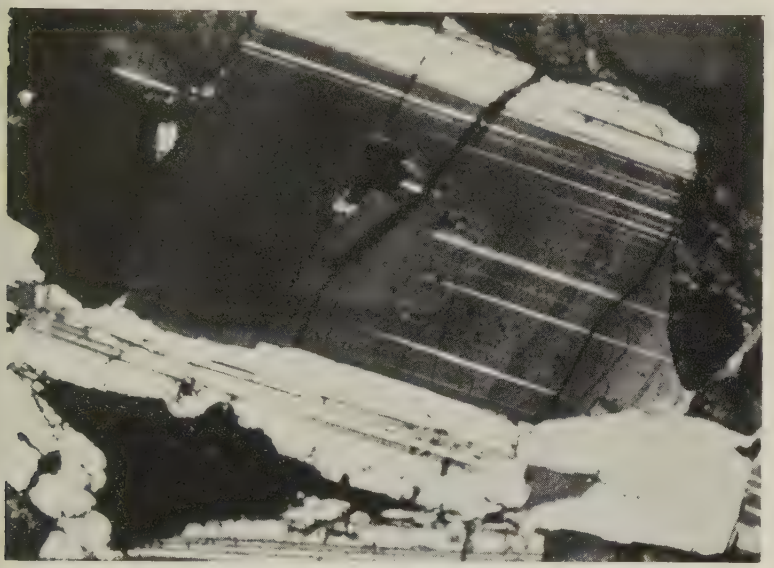

FIG. 1. A felspar crystal in hortonolite-ferrogabbro 2580. Showing primary precipitate zoning of oscillatory-normal type. The darker zones are the more calcic. Thus there are three reversals, each followed by normal zoning. The latter is strongest midway between the reversals. Size of crystal 3 mm., crossed nicols.

all the layered rocks it has the primary precipitate felspar as tabular crystals enlarged only slightly by the interprecipitate addition. The felspars, averaging 3 4 mm. in length, are fresh, and twinning, especially on the albite law, is frequent. The zoning is normal in type and results in a compositional range of about 7 parts of anorthite. The innermost parts of the crystals are An_{59} (table I and appendix). Except for an occasional patchiness, the zoning is gradual from the centre to the margin. A small proportion of the crystals fail to show zoning. Probably its absence is more apparent than real; a disadvantageous orientation to the plane of section is to be expected in some cases.

The primary precipitate zoning in the felspar of the hortonolite-

ferrogabbro 2580 is of two types. About two-thirds of the crystals are zoned in a similar fashion to those of the lower hypersthene-olivine-gabbro 4077. The remaining crystals possess a very regular oscillatory-normal zoning, as defined by Phemister (1934). There occurs a succession of reversals, up to three in number and all of a calibre estimated as 3 parts of anorthite (fig. 1). Between successive reversals is normal

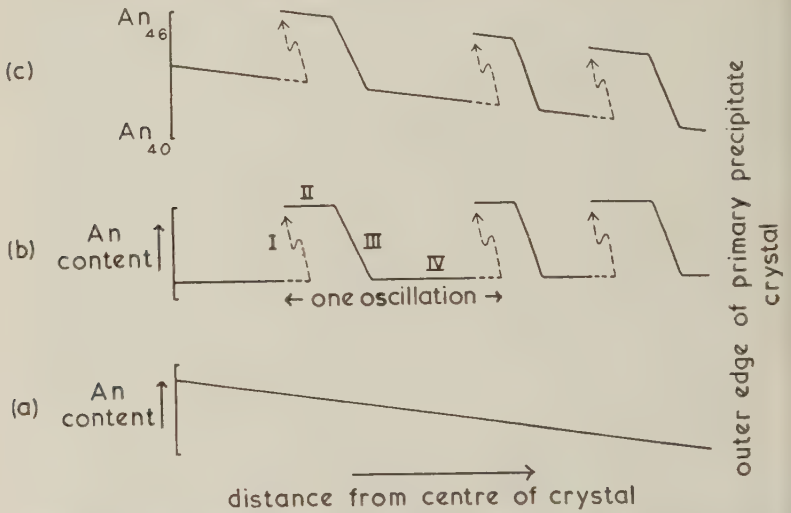

Fig. 2. Diagrams of the oscillatory zoning seen in fig. 1 and of its separate components. Broken lines and arrows denote resorption. (*a*) The simple component of normal zoning. (*b*) The oscillatory component. Roman numerals identify the successive stages of zoning. (*c*) The resultant zoning, as observed.

zoning in three well-defined stages. The first and last stages are of a zoning so gentle as to be scarcely perceptible; these are separated by an intermediate and short-lived stage of relatively strong zoning (fig. 2*c*). The composition immediately preceding each reversal is slightly more albitic than that of any material previously deposited. The boundaries of each zone are idiomorphic, though occasionally resorption prior to reversal has produced irregularities. For example, the nucleus of the crystal seen in fig. 1 is embayed by the innermost reversal, though the irregularity is poorly visible in the microphotograph.

In discussing the origin of this oscillatory-normal zoning it will be found convenient to regard it as the resultant of two components, one being oscillatory and the other simple in character. The components and the observed zoning, their resultant, are shown diagrammatically in fig. 2. The overall range in composition of an oscillatory-zoned crystal

was measured as 6 parts of anorthite (table I). Since the calibre of the reversals is estimated as 3 parts of anorthite, this overall range is contributed by the components in roughly equal proportions.

The evidence for normal zoning in the primary precipitate felspar of the highest-occurring specimen, ferrohortonolite-ferrogabbro 4145, lies in the determined range of composition, from An_{43} to An_{39} (table I). The zoning is, however, masked by the fine semi-opaque dust of included matter that prevails throughout all felspars of this specimen.

A cursory examination of a representative collection of gabbros from other horizons in the main layered series only showed oscillatory-normal zoning to affect the primary precipitate felspar of the middle gabbro 1919 (900 metres below the previously-described example).

Oscillatory zoning is usual in the felspar of the gabbros belonging to the transitional layered series. There is reason to believe, however, that these rocks differed considerably from those of the main layered series in their manner of formation. The zonal pattern of their felspars is not entirely similar and has not been fully studied. The 'perpendicular felspar rock' of the border group also possesses oscillatory-zoned felspars, but again has a specialized origin (Wager and Deer, 1939, p. 144).

III. The probable origin of the primary precipitate zoning.

In order to explain many of the outstanding structural features of the Skaergaard intrusion, Wager and Deer found it necessary to assume that a system of convective circulation was active in the magma-chamber throughout the period of formation of the main layered series (Wager and Deer, 1939, pp. 267–8). They postulated that cooled and partly-crystallized magma descended at the walls, whilst hotter magma rose along the central axis of the intrusion. Growth of the primary precipitate crystals was thought to begin in cooling magma close to the roof, to continue during descent and only to cease when the crystal was deposited on the floor and buried by others. The authors believed that the magma was effectively stirred by its convective circulation, so that temperatures in the cooler, descending currents did not differ greatly from those in the uprising magma. As a measure of the existing temperature differences Wager and Deer (1939, p. 268) accepted the estimate of Jeffreys that a thermal gradient of about 0·3° C. per kilometre should be established in convecting basalt magma.

The depth of the residual Skaergaard magma-chamber at the time of formation of the middle gabbros was about 1·5 kilometres. The available information therefore indicates the maximum difference of temperature

then prevailing in the magma to have been less than 1° C. In view of this apparent uniformity of temperature (and thus of magma composition) it is suggested that zoning was mainly caused by variation in hydrostatic pressure due to vertical movement of the convecting magma and crystals. It is considered that crystallization giving the simple normal zoning shown by the majority of crystals is the result of increasing pressure during descent. Theoretically, the effect of pressure changes on plagioclase composition is governed by the change in shape of the liquidus and solidus curves with pressure, regarding which no experimental evidence is available.[1] Existing data suffice only to afford an estimate of the increase in melting-point of the pure end-members with pressure:[2] the value dT/dP for albite is thereby indicated to be approximately four and a half times that for anorthite. Using this relationship, fig. 3 shows diagrammatically the manner in which the required change towards a more albitic solid phase might occur as pressure increases.

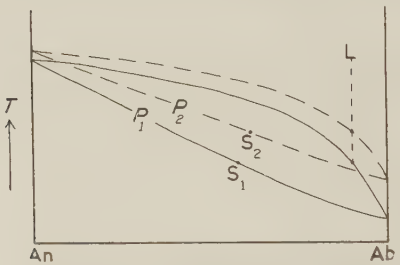

Fig. 3. Hypothetical representation of the possible effect of pressure upon the plagioclase equilibria. Solid curves represent the liquidus-solidus at atmospheric pressure (P_1). Broken curves represent the liquidus-solidus at high pressure (P_2). L is the unchanging composition of the liquid phase. With increase of pressure to P_2 the solid phase in equilibrium would change from S_1 to the more sodic composition S_2.

The simple zoning of most crystals is thus regarded as an inevitable accompaniment of the accepted process of crystallization during descent. For the oscillatory-zoned felspars, however, a more complicated growth-history must be envisaged. In the homogeneous Skaergaard magma a reversal of plagioclase zoning could only occur through a relief of pressure. It is therefore assumed that a selection of the crystals experienced

[1] The writer is indebted to H. S. Yoder, of the Geophysical Laboratory, Washington, for advice concerning this point.

[2] The Clausius-Clapeyron equation is $dT/dP = T\Delta V/\Delta H$. The values of the partial volume change ΔV (albite 0·036 c.c./gm., anorthite 0·008 c.c./gm.) were computed by H. S. Yoder from the measured densities of synthetic crystals and glass at room-temperature (Birch et al., 1942, p. 15). The enthalpies ΔH have been measured (Kracek and Neuvonen, 1952, p. 293). ΔH albite = 49·2 cal./gm., ΔH anorthite = 64·7 cal./gm. Since the values of ΔV do not strictly apply at melting temperatures, and because both ΔV and ΔH are functions of pressure, the obtained values of dT/dP are close approximations only.

a stage of ascent which led to renewed crystallization of calcic material under reduced pressure. The number of reversals present in a crystal is believed to be the same as the number of times that the crystal was carried round in the magma before finally coming to rest. That each ascent took the crystal to a similar height in the chamber is shown by the uniform calibre of all reversals (fig. 2*c*). Nevertheless, there is a constant trend of comparable and later zones towards a more albitic composition, so that the later reversals fail to achieve the earlier calcic composition. Pressure variation seems incapable of explaining this part of the zoning, which is therefore attributed to a change in the composition of the magma as overall cooling and differentiation of the intrusion proceeded. In fig. 2*a* and *b* respectively the observed zoning is resolved into this component of normal zoning and into an oscillatory component that is wholly due to pressure variation. The distinction allows the pressure effect to be studied more readily. Thus, in fig. 2*b*, a full oscillation of zoning was achieved during a complete circulation of the crystal around the magma-chamber, and its four stages correspond to successive stages in the movement of the crystal. During the first stage of a renewed circulation, namely ascent, the crystal experienced a relief of hydrostatic pressure that caused instability and resorption of its outermost sodic layers (stage I). Reversal occurred when pressure had ceased to fall and the crystal was travelling outward beneath the roof. In this high-level current the felspar suffered little or no vertical movement, so that stage II is free of zoning due to pressure change. The relatively strong zoning that follows (stage III) is comparable in origin to that affecting the simply-zoned felspars, and marks the descent stage. To complete the circulation low-level currents carried the crystal inwards to the centre of the intrusion. This final stage, being primarily one of horizontal movement, resulted in a uniformly sodic zone (stage IV). In the event of a further circulation of the crystal taking place, the zone would be partially destroyed by ensuing resorption.

Acknowledgements.—The work was carried out at Durham Colleges under the supervision of Professor L. R. Wager, whom the writer desires to thank both for the use of his material and for helpful advice.

Appendix.

Notes on the measurement of the zoning.

Values for the ranges of primary precipitate zoning are afforded by: (*a*) refractive index measurements performed on cleavage-flakes (Tsuboi,

1923); and (*b*) measurement of maximum extinction angles in the zone [010] (the zone method of Rittmann, described by Emmons, 1943). The two methods, of which the second was carried out on two of the specimens only, yield results which differ slightly at the sodic limit (table I). Whereas the zone method permits a ready distinction between primary precipitate and interprecipitate material, these two phases, when observed in the crushed state, are separable only by arbitrary means. At the time the refractive index determinations were made the prevalence of zoning in the primary precipitate felspar was not properly appreciated, and separation of the two phases was essayed by rejecting from measurement all flakes which showed zoning. The wider values obtained by the zone method seem to show that this rejection procedure was too drastic, and that it resulted in an incomplete assessment of the zoning at the sodic end. For this reason the values due to the zone method are accepted as the probable range of primary precipitate zoning.

The calcic limit of plagioclase 4077 is confirmed by a measurement of the composition of the central part of a selected crystal, using the Fedorov method.[1] The transposed poles of cleavage-planes and twin-axes gave four values of An_{59} and one each of An_{58} and An_{60}. These values agree closely with those obtained by the other methods.

No estimates of the volume and average composition of the interprecipitate felspar can be of sufficient accuracy for the determined ranges of primary precipitate zoning to be confirmed from the bulk analysed compositions.

The technique adopted for each method is described below.

(*a*) Measurement of refractive indices on cleavage-flakes. An improvised but effective temperature-variation method was used. The specially-prepared immersion liquids possessed indices successively differing by 0·001, on average. An Abbe refractometer was used in constructing dispersion-curves for all liquids. During the felspar measurements full precautions were taken to preserve accuracy. The swing-in thermometer was calibrated with that of the refractometer, and monochromatic light was used. The pure felspar was crushed and sieved to a flake-size of 0·007 mm. Because of the primary precipitate zoning the suitable flakes in a sample possessed rather variable indices. The temperature of the stage was noted when the liquid matched an index of one or more flakes. Having risen farther it was then allowed to fall and a further observation made, not necessarily on the same flakes. In

[1] Migration-curves due to Spaenhauer and reproduced by C. T. Barber (1936) were employed.

table I 'number of flakes used' means the number of such matchings. The experimental error, discounting that due to zoning, is estimated as ± 2 parts of anorthite.

(*b*) Measurement of maximum extinction angles (zone method).—A Leitz universal stage was used. Normal multiple twinning was identified as advised by Emmons (1943, p. 129). Albite-Carlsbad twins were preferred, since a maximum value was thus assured. The zone [010] was made vertical by obtaining equal illumination of the albite twin-units under the sensitive plate and with the (010) trace at 45° to the nicols. Each recorded value was verified as at a maximum and represents the mean of at least four readings. The nucleus and the margin of the primary precipitate crystal were each measured. Compositions were interpreted from the curves given by Winchell (1951, p. 262). This method of measuring plagioclase composition has been found by the writer, during an extensive investigation of a suite of felspars from Skye, to be as accurate as any other method employed on zoned felspar. The limit of error in the present case is ± 2 parts of anorthite.

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Crystallization of pyroxenes in an iron-rich diabase from Minnesota.

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THE Beaver Bay diabase forms an irregular sill-like intrusion about 3000 feet in thickness exposed over a distance of approximately six miles from Split Rock to Beaver Bay on the north shore of Lake Superior, Minnesota. It is intrusive into the Keeweenaw volcanic series above the Duluth gabbro to which it is almost certainly related. It is the largest of many such sills in this region and includes several petrographic facies grading from troctolite, through olivine-gabbro, normal gabbro, and ferrogabbro to red granite.

Analyses of several types of diabase from the Minnesota coast near Beaver Bay are given by Grout and Schwartz (1939) and include one of the iron-rich facies (table I, anal. 3) collected from Highway 61 about $1\frac{1}{4}$ miles SW. of the Beaver Bay settlement. A similar rock (table I, anal. 2) collected from the roadside about $\frac{5}{8}$ mile nearer Beaver Bay settlement and kindly supplied by Dr. F. Grout, exhibits several petrographic features worthy of comment. Tilley (1950) has drawn attention to the similarity between the fractionation in this diabase and that of the Skaergaard intrusion (Wager and Deer, 1939). For comparison a new analysis is given (table I, no. 1) of a Skaergaard ferrohortonolite-ferrogabbro showing similar iron enrichment to the Beaver Bay rock.

The analysed specimen of the Beaver Bay rock (table I, anal. 2) is a mesotype, rather coarse-grained diabase composed chiefly of clear subhedral plagioclase, rounded grains of fayalitic olivine, and three pyroxenes occurring either as stout subhedral grains up to one millimetre in length (ferroaugite) or as clusters of smaller anhedral grains composed of all three pyroxenes associated with olivine and iron-ores. Apatite, sometimes pigmented, is a prominent accessory, magnetite and ilmenite are the principal ores, and pyrrhotine is sometimes present as lamellae intergrown with the magnetite. Interstitially a little quartz is found in micrographic intergrowth with oligoclase and potash-felspar.

The plagioclase (An_{44}) occurs as subhedral grains up to two millimetres

in length which are practically unzoned, and twinned chiefly after the albite, Carlsbad, albite-Carlsbad, and pericline laws. Strong marginal

TABLE I. Rock analyses.

	1.	2.	3.	4.
SiO_2 ...	43·05	45·87	45·84	52·41
Al_2O_3 ...	10·58	10·66	11·19	12·23
Fe_2O_3 ...	4·49	4·11	4·02	3·53
FeO ...	18·23	15·59	16·30	12·86
MgO ...	3·92	3·50	3·66	2·62
CaO ...	10·37	8·28	8·39	8·48
Na_2O ...	3·20	2·84	2·68	2·01
K_2O ...	0·60	0·96	0·83	0·85
H_2O+ ...	0·11	1·42	0·87	0·92
H_2O- ...	0·16	0·47	0·45	0·14
TiO_2 ...	2·54	4·65	3·44	3·14
P_2O_5 ...	2·95	1·70	1·74	0·23
MnO ...	0·22	0·29	0·27	0·26
S ...	—	—	0·04	—
CO_2 ...	—	—	—	0·12
	100·42	100·34	99·72	99·80

Norms.

		1.	2.	3.	4.
Q		—	2·58	1·74	12·82
Or		3·34	6·12	5·00	5·28
Ab		27·25	24·10	23·06	16·87
An		12·79	13·34	15·85	21·85
Di	Wo	8·12	6·96	6·15	7·71
	En	4·00	8·70	9·20	6·55
	Fs	10·69	18·08	20·86	15·91
Ol	Fo	4·06	—	—	—
	Fa	11·42	—	—	—
Il		4·86	8·82	6·54	5·99
Mt		6·50	6·03	5·80	5·17
Ap		7·06	4·03	4·03	0·37

Normative pyroxene.

	1.	2.	3.	4.
Wo ...	35·6	20·7	17·1	25·6
En ...	17·5	25·8	25·4	21·4
Fs ...	46·9	53·5	57·5	53·0

$$\frac{FeO + Fe_2O_3 \times 100}{MgO + FeO + Fe_2O_3}$$

Ferroaugite	72	69	—	
Rock ...	86	89	86	82

1. Ferrohortonolite-ferrogabbro EG 4272. Skaergaard intrusion, east Greenland. Analyst, I. D. Muir.

2. Iron-rich diabase M3174 Highway 61. Five-eighths mile S.W. of settlement Beaver Bay, Minnesota. Analyst, J. H. Scoon.

3. Iron-rich diabase Highway 61 (Grout and Schwartz, 1939, table II, anal. 2). Analyst, R. B. Ellestad.

4. Iron-rich dolerite EH52 of New Amalfi intrusive sheet, South Africa. (Poldervaart, 1944, table VI.) Analyst, W. H. Herdsman.

zoning to oligoclase (An_{20}) is exhibited by crystals adjoining the quartzofelspathic mesostasis.

Olivine (Fa_{80}), occurring as small rounded and partly resorbed grains (table II, anal. 2), has an unusual habit for such an iron-rich member of the series. It has ceased crystallizing at a relatively early stage and is

invariably surrounded either by a reddish-brown highly-birefringent alteration product which is mostly bowlingite, or, more rarely, by deep-green isotropic chlorophaeite. Sometimes the olivine is enclosed by a pyroxene or stout grains of magnetite, but the thin mantle of bowlingite always intervenes.

Three distinct pyroxenes occur, ferroaugite, pigeonite, and hypersthene. Ferroaugite, which is the most abundant, is practically unzoned,

TABLE II. Mineral analyses.

	Mineral analyses.				Atoms on basis of 6 oxygens.		Atoms on basis of 4 oxygens.
	A.	1.	2.		A.	1.	2.
SiO_2	47·70	48·86	30·42	Si	1·886	1·906	0·968
Al_2O_3	1·55	1·51	0·50	Al	0·074	0·071	0·019
TiO_2	1·24	0·93	1·20	Ti	0·037	0·023	—
Fe_2O_3	1·82	2·26	nil	Al	—	—	—
FeO	20·32	18·71	57·62	Ti	—	0·005	0·038
MnO	0·38	0·38	n. d.	Fe‴	0·049	0·066	—
MgO	8·73	9·48	8·17	Fe″	0·672	0·608	1·526
CaO	17·32	17·58	1·32	Mn	0·012	0·014	—
Na_2O	0·65	0·24	n. d.	Mg	0·520	0·550	0·389
K_2O	0·13	nil	n. d.	Ca	0·736	0·735	0·044
H_2O+	0·21	0·25	0·18	Na	0·050	0·019	—
H_2O-	0·11	0·06	0·21	K	0·007	—	—
	100·16	100·26	99·62	z	2·00	2·00	0·99
				$X + Y$	2·05	2·00	2·00

A. Ferroaugite from ferrohortonolite-ferrogabbro EG 4272. Skaergaard intrusion. (Muir, 1951, table II, no. 14.)

1. Ferroaugite from iron-rich diabase (M3174), Beaver Bay. Analyst, I. D. Muir.

2. Olivine from same. Analyst, I. D. Muir.

and is sometimes mantled abruptly by pigeonite or hypersthene, but the reverse is also found. It is pinkish in colour, subophitic in habit, and exhibits two sets of very fine exsolution lamellae. The lamellae parallel to (100) are probably of hypersthene (Poldervaart and Hess, 1951); and the second set, extremely fine lamellae parallel to (001), have the same optical orientation as any mantling pigeonite. The ferroaugite sometimes mantles the olivine, but there is no suggestion of a reaction relationship.

Unlike the ferroaugite, the pigeonite occurs rarely as discrete crystals, but is found mostly as mantles to the lime-rich clinopyroxene or as a constituent of the clusters of ferromagnesian minerals, where it is frequently associated with the inverted orthopyroxene; both minerals may enclose olivine. It varies little in refractive index but the optic axial

angle ranges 0 -10° in different grains with the optic axial plane parallel to (010). Conspicuous exsolution lamellae of augite developed parallel to (001) are much coarser than the corresponding pigeonite lamellae found in the accompanying ferroaugite—an unusual feature.

Hypersthene (orthoferrosilite, Of_{74}), with sheets of ferroaugite inclusions parallel to (001) of the original clinopyroxene, is more abundant than pigeonite. It tends to occur mostly in the clusters of ferromagnesian crystals, but is sometimes found as large crystals mantling the ferroaugite. Where such hypersthene adjoins the late mesostasis it is frequently mantled by a primary orthopyroxene (Of_{75}) with very fine exsolution lamellae of augite developed parallel to (100). In such circumstances the orthopyroxene is not infrequently associated with a brown hornblende or intergrown with skeletal ilmenite.

Some composite pyroxene grains display relics of pigeonite enclosed in a single crystal of inverted hypersthene (fig. 1A). The parallelism of the crystals, the similarity of textures, and the identity of the bulk composition of the hypersthene (from a partial analysis) with that deduced optically for the pigeonite, indicates that most of the original pigeonite of the rock has inverted to hypersthene and that in this process coarse lamellae of ferroaugite were exsolved. These are continuous with the finer lamellae in the residual pigeonite. As most of the remaining pigeonite occurs either as small crystals intergrown with ferroaugite or as narrow mantles thereto, it would appear that the stable augite framework has tended to preserve the pigeonite, because of the parallelism and similarity of the two structures. Hess and Henderson (1949) have described a somewhat similar case where a small proportion of the pigeonite of a meteorite has inverted to orthopyroxene and the remainder exhibits very broad exsolution lamellae of augite.

From the very fine nature of the exsolution lamellae in the ferroaugite of this rock and the inversion of part only of the pigeonite, it can be inferred that cooling in the Beaver Bay sill was more rapid than that of comparable rocks of the Skaergaard intrusion where exsolution lamellae of pigeonite in the ferroaugite are much coarser. Hypersthene lamellae parallel to (100) are seen only in the early magnesian clinopyroxenes from the Skaergaard hypersthene-olivine-gabbros. In the Skaergaard clinopyroxenes of the middle and ferrogabbros, exsolution took place while the temperature was still above the pigeonite-orthopyroxene inversion, but in the Beaver Bay rocks with more rapid cooling only a little pigeonite was able to exsolve above the inversion temperature and the remaining excess of the lime-poor phase has separated as

hypersthene lamellae parallel to (100). Optical data for these minerals are listed in table III.

The closest chemical comparison with the two analyses of iron-rich Beaver Bay diabases is a ferrohortonolite-ferrogabbro (table I, anal. 1)

Fig. 1. Iron-rich diabase of Beaver Bay, Minnesota.

A. Composite grain in centre of field shows a resorbed crystal of fayalitic olivine altered marginally to bowlingite, enclosed in a large crystal of inverted orthopyroxene with some of the original pigeonite remaining. On the left side of the field a little ferroaugite mantles the olivine. Iron-ore and pigmented apatite are prominent accessories.

B. i. (upper left).—Resorbed olivine with bowlingite mantle enclosed by pigeonite intergrown with ilmenite. The pigeonite in turn is enclosed by ferroaugite. ii. (upper right).—Ferroaugite with feeble exsolution lamellae parallel to (001) is margined by a narrow rim of pigeonite showing conspicuous exsolution lamellae. Olivine on extreme right has a broad mantle of inverted orthopyroxene enclosing relics of pigeonite. Inverted hypersthene in the upper part of the field has a clear margin of primary hypersthene adjacent to the quartzo-felspathic mesostasis. Much of the late plagioclase is antiperthitic. iii. (lower portion).—Large crystal of ferroaugite and a smaller grain of pigeonite, part of which has inverted to hypersthene with sheets of clinopyroxene inclusions. The pyroxenes are set in a base of bowlingite derived from the olivine.

of the Skaergaard intrusion. As the Beaver Bay rocks are considerably poorer in lime than the Skaergaard ferrogabbros and carry a more basic plagioclase, their normative pyroxenes are much poorer in lime and pigeonites are present in the iron-rich facies, whereas in the Skaergaard intrusion, lime-poor pyroxenes have ceased to form much above the base of the ferrogabbros. Like the analysed Skaergaard rock, the iron-rich Beaver Bay diabases are very high in phosphorus. Analyses of more magnesian types from the intrusion are very low in this constituent

TABLE III. Optical properties and compositions of pyroxenes and associated olivines.

	1.	2.	3.	4.	5.	6.	7.	8.	9.
α	1·718	—	1·704	1·729	1·741	1·787	1·711	—	—
β	1·725	1·818	1·711	1·730	1·750	1·821	1·716	—	—
γ	1·741	—	1·733	1·757	1·757	1·838	1·738	—	—
2V	52°	58°	50°	0–10°	65°	58–56°	46–56° av. 49°	64–67° av. 66·5°	54·5°
$\gamma:c$	43°	—	39°	40°	—	—	43°	—	—
Colour ...	brownish	pale yellow	brownish	pale yellow-green	pale green	pale yellow	brownish	pale green	pale yellow
Pleochroism	feeble	none	feeble	feeble	feeble	none	feeble	feeble	none
Composition atomic % Ca	37	—	38	9	—	2	37	—	—
Composition atomic % Mg	26	22	28	27	25	20	27	27	15
Composition atomic % Fe	37	78	34	64	75	78	36	73	85

Values of all refractive indices $\pm 0{\cdot}002$. 2V and extinction angles $\gamma:c \pm 2°$.

1. Ferroaugite from ferrohortonolite-ferrogabbro EG 4272. (Table I, anal. 1.) Skaergaard intrusion.
2. Olivine from same.
3. Ferroaugite from iron-rich diabase M3174 Beaver Bay. (Table I, anal. 2.)
4. Pigeonite from same.
5. Hypersthene from same.
6. Olivine from same.
7. Ferroaugite from iron-rich dolerite EH52. (Table I, anal. 4.) New Amalfi intrusive sheet, South Africa.
8. Hypersthene from same.
9. Olivine from same.

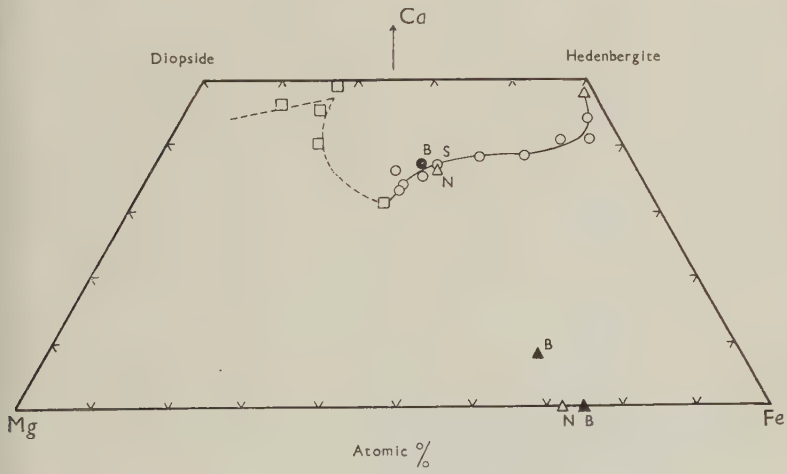

FIG. 2. Pyroxenes from the Beaver Bay diabase.

Comparison is made with the Skaergaard normal clinopyroxene trend (Muir, 1951) and with the pyroxenes in an iron-rich dolerite from New Amalfi, South Africa.

○ ● Analysed pyroxenes. □ Clinopyroxene composition recalculated from analysis of bulk pyroxene of the rock. ▲ △ Composition estimated from optical properties. B = Beaver Bay pyroxenes. N – New Amalfi pyroxenes. S = Skaergaard ferroaugite (table II, anal. 1).

and it would appear that here as with the Skaergaard example the phosphorus content of the magma increased until it was saturated and then apatite appeared suddenly as a major constituent.

The compositions of the ferromagnesian silicate minerals of the Beaver Bay and Skaergaard rocks are plotted in fig. 2 along with those from a comparable iron-rich dolerite of the New Amalfi intrusive sheet, South Africa (Poldervaart, 1944). From this, and the data of table IV, it can be seen that the distribution of iron and magnesium between the different associated minerals follows a generally similar pattern although the olivine of the Beaver Bay diabase is of early formation, whereas in the other cases it formed after the ferroaugite. It is also of interest to note that both the Beaver Bay and New Amalfi ferroaugites lie on the Skaergaard trend (Muir, 1951).

Table IV. Iron enrichment in associated ferromagnesian minerals.

	$\frac{Fe}{Fe+Mg}$ atomic %.		
	1.	2.	3.
Augite	59	55	57
Pigeonite	—	71	—
Hypersthene	—	75	73
Olivine	78	79·5	85

1. Skaergaard ferrogabbro (table I, anal. 1).
2. Beaver Bay diabase (table I, anal. 2).
3. New Amalfi iron-rich dolerite (table I, anal. 4).

The compositions of associated orthopyroxenes, pigeonites, and augites taken mostly from Kuno's work are plotted in fig. 3. From this it can be seen that the usual relationship is for the pigeonite to be slightly more iron-rich than its associated orthopyroxene. Using the data for the analysed pyroxene pair H_2–H_3 in the diagram, Kuno and Nagashima (1952) have been able to deduce the magnitude of the pigeonite–orthopyroxene inversion interval at one point. The reversal of the usual relationship in the case of the Beaver Bay rock supports the textural evidence that this primary hypersthene formed later and at a lower temperature than the pigeonite.

It is also of interest to compare the relative iron-enrichment of the associated orthopyroxenes and olivines. The data of table IV agree with the work of Bowen and Schairer (1935) who found that in synthetic pairs the olivine was always the more iron-rich, a relationship which was the reverse of what commonly obtains in igneous rocks. They attributed the difference to the order of crystallization of the different minerals in

basic igneous rocks, where a magnesian olivine when present is usually the earliest mineral to separate.

Ramberg and De Vore (1951) have collected data on natural olivine-orthopyroxene pairs and shown that the iron-rich minerals conform to the synthetic pattern, but that for their Wyoming syenites the difference

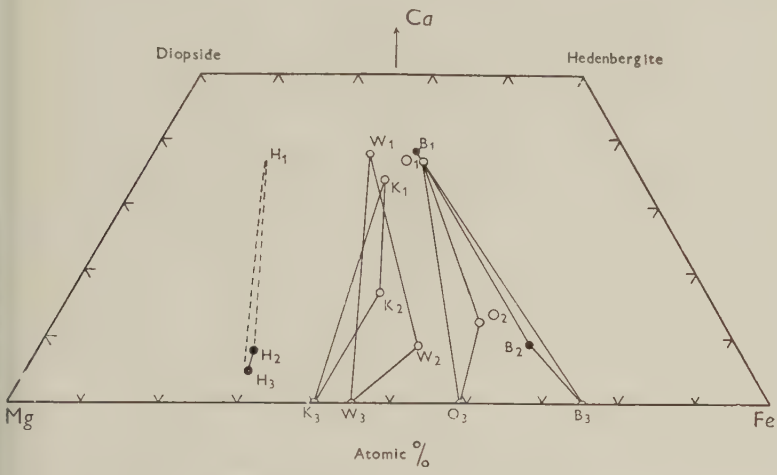

FIG. 3. Associated pyroxenes from 3-pyroxene rocks.

B. Beaver Bay diabase: B_1 ferroaugite (anal.), B_2 pigeonite, B_3 primary orthopyroxene.

H. Hakone andesite (Kuno and Nagashima, 1952): H_2 pigeonite (anal.), H_3 orthopyroxene (anal.), H_1 inferred composition of associated augite.

K. Hakone andesite (Kuno, 1936. Jap. Journ. Geol. Geogr., vol. 13, p. 125): K_1 augite, K_2 pigeonite, K_3 orthopyroxene.

O. Okuba Yama andesite (Kuno and Inoue, 1949. Proc. Jap. Acad., vol. 25, no. 4, pp. 128–132): O_1 ferroaugite, O_2 pigeonite, O_3 orthopyroxene.

W. Weiselbergite (Kuno, 1947. Proc. Jap. Acad., vol. 23, no. 9, p. 111): W_1 augite, W_2 pigeonite, W_3 orthopyroxene.

in iron-enrichment is greater on the average than for synthetic pairs. Within the magnesian range, however, they confirmed Bowen and Schairer's observations. Later, Ramberg (1952) discussed on theoretical grounds the distribution of iron and magnesium in coexisting olivines and orthopyroxenes and considered that the relative enrichment in iron shown by the olivines could be attributed to the lower average electronegativity of the oxygen forming the metal–oxygen bonds in the orthosilicate. The presence of more tetrahedrally co-ordinated aluminium in an augite would on this theory make it more receptive to

iron-enrichment than an associated orthopyroxene, but the reverse is always the case.

The two-pyroxene boundary.

All three rocks whose pyroxenes are plotted in fig. 2 appear to have formed from liquids lying close to the two-pyroxene boundary. In the case of the Skaergaard rocks, lime-poor pyroxene ceased to form only slightly later than the reappearance of olivine, and there is no evidence of a reaction relation with ferriferous olivine being responsible for the final disappearance of this pyroxene (Muir, 1951). In the Beaver Bay rocks, olivine of early formation is mantled frequently by pigeonite or its inverted equivalent. In the New Amalfi rock both the olivine and orthopyroxene have formed about the same time at a very late stage and there is no evidence of a reaction relation between them. It would appear, therefore, that the disappearance of the lime-poor pyroxene in iron-rich tholeiitic rocks could not be due to a reaction relation with fayaltic olivine.

Another suggestion which has been advanced to account for the presence of only one pyroxene (ferroaugite) in the later stages, in place of the two which are found in the earlier rocks of some intrusions, is that when the two-pyroxene boundary is approached, the binary cotectic relationship of limited solid solution between augite and pigeonite develops into a solid-solution series with a minimum and the augite and pigeonite compositions converge, one becoming more lime-poor and the other richer in lime. However, the ferroaugites associated with pigeonite are now known to become progressively more lime-rich, and this does not conform well to a change over to a complete solid-solution relationship with a minimum.

If neither of these hypotheses are accepted it must follow that minerals of the pigeonite series should continue to form as long as the composition of the magma permitted and that any disappearance of the lime-poor pyroxene phase would be due to some change in the composition of the magma towards the lime-rich side of the pyroxene field.

A new hypothesis advanced here to explain the rather complex pyroxene relations near the two-pyroxene boundary can best be illustrated by a series of hypothetical phase diagrams (fig. 4) at different stages of iron-enrichment across the pyroxene field. For this purpose the simple binary diagram alone is rather misleading because the liquidus will always be enriched in iron with respect to the solidus and

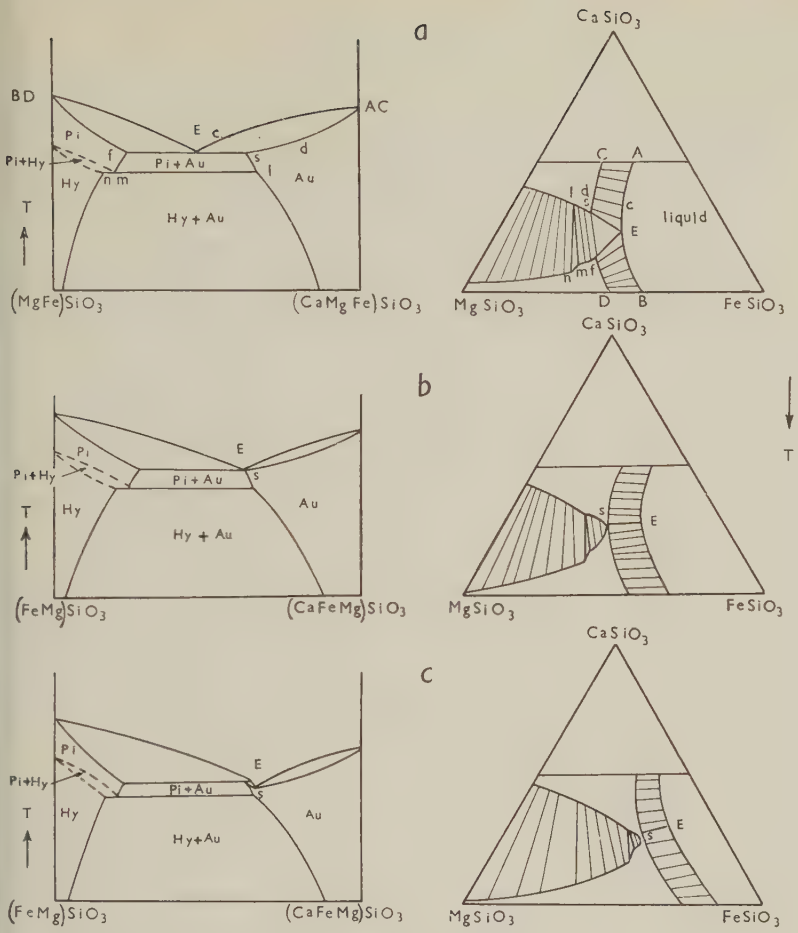

FIG. 4. Hypothetical equilibrium diagrams for pyroxenes.

The diagrams on the left represent the phase relations in a simplified manner as a pseudo-binary system. Diagrams on the right show hypothetical phase equilibria for a series of isothermal planes at successively lower temperatures and at different stages of iron-enrichment.

(*a*) Two-pyroxene field. The diagram on the right represents an isothermal plane at the temperature of the line *fEs* of the left-hand diagram. From a liquid of composition *c*, augite *d* would crystallize. If the liquid has a composition *E* on the pyroxene cotectic curve, two pyroxenes *f* (pigeonite) and *s* (augite) should separate. On the right-hand diagram the triangle *lmn* represents the pigeonite–hypersthene inversion-interval. Comparable points in the two diagrams are designated by the same letter.

(*b*) Limit of two-pyroxene field. The cotectic curve at *E* has migrated to the lime-rich side of the intersection with the solvus. From liquid *E*, a single pyroxene *s* of similar lime-content will separate. Right-hand diagram represents the isothermal plane of temperature *Es* on left-hand diagram.

(*c*) One-pyroxene field. The cotectic curve at *E* has become detached from the solvus, and ferroaugite *s* alone should separate from the liquid. Right-hand diagram represents the isothermal plane of temperature *Es* on left-hand diagram.

the two coexisting pyroxene phases will have different iron-magnesium ratios. The diagrams used to illustrate pyroxene relations in the central part of the system include a set showing relations for a series of isothermal planes with falling temperature.

The subsolidus relations for the system $Mg_2Si_2O_6$–$CaMgSi_2O_6$ have been determined by Atlas (1952) who has found that the maximum of the solvus appears to lie only very slightly below the solidus located by Bowen (1914). Thus, in the synthetic dry system, there is a simple solid-solution series with a minimum, but any addition of a little water or other constituents to the liquid would almost certainly lower the minimum, enabling the solvus to cut the solidus and produce two immiscible series of pyroxenes. With iron entering the system the maximum of the solvus should not fall as rapidly as the liquidus, and two pyroxenes would continue to separate (fig. 4*a*).

As the limit of the two-pyroxene field is approached it seems probable that the minimum of the liquidus would migrate towards the lime-rich side of its intersection with the solvus (fig. 4*b*). This should cause the proportion of lime-poor pyroxene to diminish very gradually as seems to be the case in the latest two-pyroxene rocks of the Skaergaard intrusion. When the minimum passes beyond the limit of the intersection with the solvus (fig. 4*c*) the lime-poor pyroxene should cease to separate as a separate phase, although it could still appear as exsolution lamellae in the ferroaugite on slow cooling. In different magmas the position and temperature of the minimum would vary somewhat according to the amounts of other constituents in the magma, and hence the limit of the two-pyroxene field in different magmas could occur at slightly differing iron-magnesium ratios.

Such a hypothesis could account for the disappearance of pigeonite in iron-rich tholeiitic rocks, but in some of these a primary hypersthene is present in the late crystallizing material along with augite and iron-ore. From the presence of exsolved hypersthene in the ferroaugite of the Beaver Bay diabase and of a primary hypersthene in the late crystallizing material it may be inferred that the bulk of the rock formed at a temperature not greatly in excess of the hypersthene–pigeonite inversion. An increased content of volatiles in the iron-enriched interstitial liquid could lower the crystallization temperature of the minimum below that of the hypersthene–pigeonite inversion. For rocks which have formed from liquids just beyond the limit of the two-pyroxene field (fig. 4*c*) the minimum could now once again intersect the solvus, and result in two pyroxenes (ferroaugite and hypersthene) appearing again

in the residuum. This seems to be the explanation of the hypersthene in the New Amalfi rock.

Strong zoning of moderately iron-rich augites towards subcalcic augite and lime-rich pigeonite [optic axial plane (010)] has been reported from some of the Karroo dolerites (Walker and Poldervaart, 1949), Central Australian dolerites (Wilson, 1948), NE. Otago tholeiites (Benson, 1944), and Japanese andesites (Kuno, 1936). This type of zoning is relatively uncommon and in this case it would appear that the pyroxene cotectic curve has migrated towards the lime-poor side of its intersection with the solvus and that crystallization must have been effected at rather elevated temperatures. Here then, at a medium stage of iron-enrichment, it seems that alternative trend lines are possible for tholeiitic pyroxenes, one type leading towards the relatively lime-rich ferroaugites and the other to pigeonitic types. For more extremely iron-enriched liquids the pyroxene relations may be complicated by the formation initially of iron-wollastonites such as were recognized in the Skaergaard intrusion by Wager and Deer (1939), and which, perhaps also, may have formed in the most iron-rich rocks of the New Amalfi intrusive sheet.

The author wishes to express his gratitude to Professor C. E. Tilley for drawing his attention to the problems presented by the Beaver Bay pyroxenes, and for advice and criticism of this paper. To Professor J. B. Thomson also thanks are due for helpful discussions on pyroxene problems. Professors F. Walker and A. Poldervaart very kindly supplied specimens of the New Amalfi rocks, and Mr. J. H. Scoon carried out an analysis of the diabase specimen from Beaver Bay which is published here by courtesy of Professor Tilley.

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Veatchite from the Permian evaporites of Yorkshire.

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SOME years ago a few small grains of an unknown mineral were separated by F. H. Stewart from well cuttings from the Permian lower evaporite bed of the Eskdale no. 2 borehole, sunk by the D'Arcy Exploration Company near Aislaby, north Yorkshire. The mineral (not then identified) was mentioned in an account of the mineralogy of this evaporite bed (Stewart, 1949, p. 626). Flame tests suggested that boron and strontium were present, and this was confirmed by Dr. R. L. Mitchell of the Macaulay Institute for Soil Research in Aberdeen, who, by spectrographic work, found that these were the main constituents, and that very much smaller quantities of Ca, Mg, Ba, Na, and K were present.

The mineral appeared to be a strontium borate, and was found to be optically similar to veatchite, which was then considered to be a calcium borate (Switzer, 1938). When Switzer and Brannock (1950) showed that veatchite was actually a strontium borate (the only natural one known) it became apparent that the Yorkshire mineral was identical with veatchite. The identification has been confirmed by a microchemical analysis by R. A. Chalmers and X-ray work by R. Phillips.

Physical and optical characters.

The mineral occurs in Yorkshire as colourless transparent crystals of acicular and platy habits. The needles occasionally reach 3 mm. in length and 0·5 mm. in thickness. The plates (flattened on (010)) reach 2 mm. in maximum width and 0·5 mm. in thickness. Some of the crystals show parallel growth. Several prism, dome, and ill-developed pyramid faces can be seen, but all these faces are striated and dull, and no reasonable reflections were obtained on the goniometer. Several of the crystals are doubly terminated.

There are three cleavages—perfect (010), good (001), fair (100). The (100) cleavage has not previously been recorded. The angle from the

(001) cleavage to the prism zone (59°) as measured microscopically, agrees with that of Murdoch (1939). The lustre is pearly on (010) and vitreous elsewhere. Specific gravity, determined by suspension in bromoform-benzene mixtures, is 2·6, which is in general agreement with the value 2·58 given by Murdoch, but is lower than that (2·69) given by Switzer. The mineral does not fluoresce in ultra-violet light.

One crystal was found to contain small euhedral rectangular crystals of anhydrite. Otherwise the veatchite grains contain no visible impurity.

The optical properties are: α 1·551, β 1·553, γ 1·621 ($\pm$0·002) for sodium-light; Positive 2V 37°; $\beta = b$; $\gamma : c = -30°$. These agree with those of Switzer (1938), with the exception of the angle $\gamma : c$, which he gives as $-38°$.

Chemical characters.

A microchemical analysis of hand-picked material has been made by R. A. Chalmers, using 0·013 gm. of material. Two determinations of each of the three major constituents were made. The results are quoted in table I, with analyses of Californian veatchite for comparison. From chromatographic tests it was estimated that not more than 0·5 % of CaO is present. Spectrographic work also shows that Ca is present in very small quantity.

From the American analyses, Switzer and Brannock (1950) give two possible formulae—$3(Sr,Ca)O.8B_2O_3.5H_2O$ or $SrO.3B_2O_3.2H_2O$. The Yorkshire analysis agrees closely with the second of these, the atomic ratios to 12 oxygens being $Sr_{1.01}B_{6.00}H_{4.03}$, with a small quantity of Ca replacing Sr. The first formula would not be satisfied even if the maximum indeterminate errors quoted below by Chalmers were accepted. The first formula is the one given tentatively in Dana's System of Mineralogy, 7th edition, 1951, vol. 2.

X-ray data.

X-ray powder photographs of material from Lang and from Yorkshire have been compared and found to be similar. Powder data in kX units for an acicular crystal from Yorkshire is given in table II.

Occurrence.

Veatchite has previously been recorded from only one locality, at Lang, California, where it occurs in very small quantity in boulders on the dump of an old colemanite mine. It is a constituent of thin cross-fibre veins cutting limestone and howlite (Switzer, 1938), and also forms

TABLE I. Chemical analyses of veatchite.

	1.	2.	3.	4.	5.
SrO	29·9 30·1 } 30·0	29·8	30·3	29·7	31·20
CaO	—	—	1·4	1·7	
B_2O_3	60·3 60·3 } 60·3	59·8	58·6	57·8	59·26
H_2O	10·4 10·5 } 10·5	10·4	9·7	—	9·54
	100·8	100·0	100·0	—	100·0

1. Veatchite from Eskdale no. 2 borehole, north Yorkshire.* Figure for SrO includes not more than 0·5 % CaO. Analyst, R. A. Chalmers.

2. The above analysis recalculated to 100 %.

3. Veatchite from Lang, California, recalculated to 100 % (original total 98·4). Analyst, W. W. Brannock (Switzer and Brannock, 1950).

4. Partial analysis of veatchite from Lang, California, recalculated to correct for 1 % insoluble material. Analyst, W. W. Brannock (Switzer & Brannock, 1950).

5. Veatchite from Lang, California, recalculated to 100 % (original total 100·23). Analyst, F. A. Gonyer (Switzer, 1938).

TABLE II. X-ray spacings for veatchite.

d.	I/I_0.	*d.*	I/I_0.	*d.*	I/I_0.	*d.*	I/I_0.
10·3	vs[1]	2·69	vvw	1·832	vvw	1·311	vvw
5·64	m	2·59	s[3]	1·797	vw	1·287	vvw
5·12	m	2·52	vw	1·757	w b	1·270	vw
4·46	w	2·39	m	1·673	vvw	1·247	vvw
4·00	m	2·20	m	1·650	vvw	1·225	vw
3·47	w	2·147	w	1·583	vw b	1·167	vvw
3·32	vs[2]	2·078	m	1·534	vw	1·113	vvw
3·19	w	2·033	m	1·506	vw	1·086	vw
2·98	w b	1·992	vw	1·426	vvw	1·064	vvw
2·86	m	1·932	m	1·399	vvw	1·046	vw
2·76	vvw	1·859	vw	1·353	vw		

Order of intensities: vs[1], vs[2], s[3], m, w, vw, vvw. b indicates broad line. 9 cm. camera cut off at 10·8 kX. Cu-$K\alpha$ radiation (λ = 1·5374 kX).

* Methods of analysis:

Water.—Determined once by the Penfield method, and once indirectly as loss on ignition at a temperature not sufficiently high to cause loss of boric oxide. The indeterminate error was computed to be about $\pm$0·3 % H_2O.

Strontia.—Determined once directly, by precipitation as the oxalate and subsequent titration of the oxalate with ceric sulphate; once indirectly, after the indirect water determination, by removing boron by volatilization with hydrofluoric and sulphuric acids, and final weighing of the strontium as the sulphate. The error was computed to be about $\pm$0·2 % SrO.

Boric oxide.—Determined twice by the same method. The mineral was fused with sodium carbonate, and the melt cooled and dissolved in hydrochloric acid. The solution was titrated with caustic soda to a known pH in the range 7·3 to 7·6. Mannitol was added, and the solution was re-titrated to the same pH, measured by a pH meter. The caustic soda was standardized empirically with boric acid under similar conditions. The error was computed to be about $\pm$0·5 % B_2O_3.

platy aggregates lining irregular fractures in clay (Murdoch and Webb, 1940). It is associated with colemanite (Murdoch, 1939; Switzer and Brannock, 1950).

In the Eskdale no. 2 borehole in Yorkshire, the mineral has been found in well cuttings from the following depths: 4360, 4365, 4380, 4385, 4390, 4430, 4435, 4445, 4480, 4500, 4505, 4550 feet below surface. These depths include parts of the polyhalite, upper anhydrite, and upper halite-anhydrite zones of the lower evaporite bed (Stewart, 1949). Several of the crystals are complete, and it is possible that they were enclosed in halite, since all other minerals of this evaporite bed tend to be euhedral towards halite.

This provides the only record of veatchite in evaporites of marine origin. It is the first record of a borate in this lower evaporite bed, but an analysis of a polyhalite-kieserite-rock from the Eskdale no. 3 borehole at this horizon in Yorkshire shows Sr 0·9 and B 0·02 % (Armstrong, Dunham, and others, 1951). Boracite has been recorded from the middle evaporite bed in the Eskdale no. 2 borehole (Guppy, 1944). Celestine has been recorded as a rare constituent in the Eskdale no. 2 borehole (Stewart, 1949).

Acknowledgements.—The authors would like to express their gratitude to Dr. W. T. Schaller and Dr. G. Switzer for providing specimens of veatchite from Lang; to Dr. R. L. Mitchell for his spectrographic work on the Yorkshire veatchite; and to the D'Arcy Exploration Company Ltd. (which is a fully-owned subsidiary of the Anglo-Iranian Oil Company Ltd.) for supplying well cuttings from their borehole.

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Two arfvedsonitic rhyolite intrusions from Cloghaneely, Co. Donegal.

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[Read March 25, 1954.]

Introduction.

MANY of the Dalradian and 'Moine' rocks of Donegal, and the Caledonian granites associated with them, have been shown to bear structural and petrographic resemblance to the rocks of similar age in Scotland. In Scotland riebeckite-bearing minor intrusions occur at many localities and include Ordovician, Caledonian, Carboniferous, and Tertiary intrusions. There appears to be no record of analogous rocks containing sodic amphiboles occurring in situ in NW. Ireland. The purpose of this paper is to record the existence of two small intrusions of such rocks in western Cloghaneely, Co. Donegal.

The area in which these rocks occur, although rising to over 1400 feet O.D., is thickly mantled by both glacial boulder-sand and peat bog. Consequently exposures are scarce, but sufficient outcrops occur to show that the country-rock comprises a massive lineated quartz-schist of Dalradian age. Both this quartzite and the contained metadolerite sills have sustained the same regional metamorphism. Fig. 1 shows that the Dalradian rocks have been invaded by the Donegal granite which is almost certainly Caledonian. Although numerous microgranitic and rhyolitic dikes cut the granite in various areas, none has been found with strongly sodic affinities, and surprisingly few appear to intrude the quartzitic country-rock. This suite of acid dikes was emplaced shortly after the formation of the main granite. The only later solid rocks in the area comprise a swarm of Tertiary basaltic dikes which cut the granitic, microgranitic, and Dalradian rocks. Considerably to the east McCall (1953) has recorded a suite of lamprophyric dikes, but no members of this suite occur in the present area.

The sodic rhyolite intrusions.

On the high plateau linking the summits of Tievealehid and Carntreena, numerous scaly or discoidal porcellaneous erratics are to be found. They

occur on top of the drift immediately under the bog, being seen where the latter has been 'hagged' away, and can only be matched by the rocks occurring in two small dikes. However, the abundance of the small erratics suggests that there are probably further intrusions obscured by

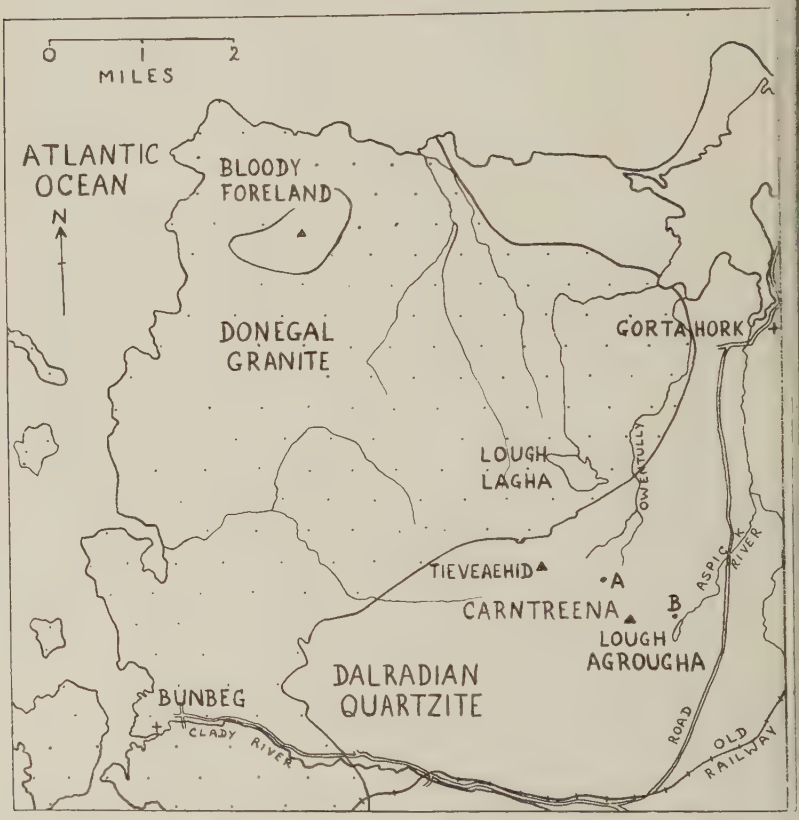

Fig. 1. Sketch-map of the Cloghaneely area, marking exposures A and B. [For Tieveaehid on the map read Tievealehid.]

the drift mantle. The two exposures are too small to determine the exact trend, but the dikes appear to run roughly N.-S.; they are:

A. NW. of summit of Carntreena, at about 1150 feet above sea level, in a gully draining down into the long eastern tributary of the Owentully. Occurs on SE. margin of a large metadolerite mass.

B. East of summit of Carntreena just NW. of Lough Agrougha, the

outcrop being 200 yards up the stream (unmarked on 6-inch map) joining the Aspick river where it leaves the lough.

Petrology of the two intrusions.

Intrusion A.—Hand-specimens appear to be perfectly homogeneous with a general appearance of impure chalk (despite the superior hardness) due to the off-white colour. In thin section a very strong fluxion structure is revealed in a nearly monomineralic albitic rock. These felspar laths range up to 0·4 mm. long, whilst occasional more stocky microphenocrysts (0·5 mm. wide) of albite occur. These felspars are considerably decomposed. Subhedral and euhedral sodic amphiboles (about 0·1 mm. long) are scattered through the rock with a preferred orientation of the *c*-axes parallel to the fluxion structure. Very rare corroded amphibole phenocrysts up to about 0·9 mm. length have been detected. The extreme rarity of these phenocrysts makes complete optical determination difficult, but they appear to be ordinary common hornblende with α straw-yellow, β straw-yellow, γ rich leaf-green; $\gamma > \beta \geqslant \alpha$; $\beta = b$; optically negative.

These hornblendes are very highly corroded, giving ragged margins studded with abundant magnetite grains, which to some extent show the original shapes of the grains. One grain also showed the whole central part of the crystal 'scoured' and replaced by ordinary groundmass constituents. These amphiboles are probably xenocrystic. Yellow-green epidote occurs as very irregular minute prisms, which frequently possess dusky-brown cores. Though much smaller, the epidotes are often numerically as abundant as the sodic amphiboles. Very small amounts of quartz occur interstitially.

Intrusion B.- Hand-specimens are generally similar to those of intrusion A, except for little clusters of black spots. The dominant minerals are albite (Ab_{91-95}) dusted with secondary 'white mica', and quartz, in a volumetric ratio of about 3:1. The grains are more equidimensional and with less marked fluxional texture than in intrusion A. The albite forms larger and more euhedral grains (average length 0·1 mm., and sometimes reaching 0·2 mm.) than the quartz (average size 0·05 mm., rising to 0·1 mm.). The macroscopic black spots prove to be large sodic amphiboles up to 2 mm. across; these are always extraordinarily sieved, giving clusters of apparently isolated fragments which are never more than about 0·25 mm. in diameter. Although in optical continuity, each fragment appears to be interstitial to the quartzo-

felspathic groundmass. Minute grains (up to 0·045 mm.) of interstitial yellow-green epidote occasionally occur.

The sodic amphibole.

In both rocks the sodic amphibole bears a superficial resemblance to riebeckite, but closer optical examination discloses somewhat complex relationships, and shows that the minerals are closer to arfvedsonite than riebeckite.

In intrusion A the sodic amphibole has been found to possess two different types of optical orientation. The rather less common type (i) shows:

α fairly deep blue-green, β pale orange-yellow, γ dirty bluish- or greenish-yellow; $\alpha > \gamma > \beta$.

and the more abundant orientation-type (ii) shows:

α orange-yellow to pale watery olive-green, β dirty greeny-yellow, γ fairly dense olive-green, slightly blued; $\gamma > \beta > \alpha$.

The other optical characters are as follows:

Type (i).	Type (ii).
2V, negative, very small	2V, positive, very small, almost uniaxial
$\gamma = b$; op. ax. pl. $\perp$ (010)	$\beta = b$; op. ax. pl. $\parallel$ (010)
negative elongation	positive elongation
$\alpha : c$ = about $> 10°$	$\gamma : c$ = about 17°
extremely small birefringence	very small birefringence
strong dispersion, anomalous extinction	
Size: larger prisms about 0·1 mm. long	

Type (i) possesses the standard arfvedsonite orientation, but in the case of type (ii) there is at first sight a considerable discrepancy. A very slight variation in composition would undoubtedly cause minor alteration of the refractive indices, and since the birefringence of type (i) is extremely small, the sign and position of the optic axial plane could easily alter. Since the birefringence is only of the order of 0·002, it would only necessitate, for example, a reduction of γ by 0·003 to give a complete re-orientation in which α, β, and γ of (i) would become γ, α and β: and, instead of having $\gamma - b$ and negative sign, would now have $\beta - b$ with positive sign. Since type (ii) bears just this sort of relationship to type (i), it seems reasonable that both should be termed arfvedsonite. It is clear from the absorption formulae given above that, but for the re-orientation, both types show very analogous pleochroism. Microscopically they show similar habits and textural relationships, and it thus seems probable that these two types represent amphibole whose

composition is slightly variable, possibly due to slightly different times of crystallization.

Data on the optical properties of arfvedsonite show quite considerable variation, but Sundius (1946) suggests that the strong absorption colours and low birefringence are characteristic, whilst a review of recently

TABLE I. Optical data for sodic amphiboles.

	Sign.	Optic axial plane.*	$\alpha:c$.	$\gamma-\alpha$.	2V.	Pleochroism α.	Pleochroism β.	Pleochroism γ.
					ARFVEDSONITE			
1.	—	—	−14 to 20°	—	—	—	—	—
2.	—	—	small	small	—	intense bluish-green to indigo	greyish-violet	light yellowish-green
3.	neg.	⊥	10	0·005	—	dark bluish-green	brownish-yellow	dark bluish-grey
4.	neg.	⊥	15	0·005	—			
5.	neg.	⊥	10	0·005	—			
6.	pos.	∥	—	0·026	64 to 73°	dark bluish-green	greenish or greyish-brown $\alpha > \beta > \gamma$	yellowish-brown or greyish-yellow
7.	neg.	⊥	8	0·003†	large	—	—	—
8.	neg.	⊥	15	0·004†	—	—	—	—
9.	—	—	30–35	—	—	—	—	—
10.	—	—	8	—	—	blue	indigo-blue $\beta \geqslant \alpha > \gamma$	yellow-green to brownish-green
11.	neg.	—	44‡	0·009	—	pleochroic in light and dark yellow		
12.	neg.	⊥	> 10	extremely small	very small	see p. 396 $\alpha > \gamma > \beta$		
13.	pos.	∥	~ 17‡	very small	almost uniaxial	see p. 396 $\gamma > \beta > \alpha$		
14.	neg.	∥	> 10	~ 0·013	~ 45°	see p. 397 $\alpha > \beta > \gamma$		
					KATAPHORITE			
15.	neg.	—	36	0·007†	—	—	—	-
16.	neg.	—	56	0·007†	small	—	—	—
					ECKERMANNITE			
17.	neg.	—	25–53	0·013	75°	—	—	—

* Optic axial plane perpendicular or parallel to (010).

† $\beta-\alpha$ instead of $\gamma-\alpha$. ‡ $\gamma:c$ instead of $\alpha:c$.

1, Johnsen, 1901. 2, De Angelis, 1923. 3–5, Gordon, 1927. 6, Kupletsky, 1927. 7–8, Kunitz, 1930. 9, Sobolev, 1933. 10, Coulson, 1936. 11, Wahlstrom, 1940. 12–14, Whitten, 1954. 15–16, Kunitz, 1930. 17, Sundius, 1946.

described arfvedsonites shows that the pleochroism scheme is usually differentiable from that of riebeckitic types (see table I). The strongly sieved grains from intrusion B give consistent optics, as follows:

α opaque inky prussian-blue, β deep greeny-blue, γ grass-green to yellow-green; $\alpha > \beta > \gamma$
2V, negative, about 45°; $\beta = b$ with negative elongation, $\alpha:c > 10°$
$\gamma-\alpha$ appears to be at least 0·013
Practically all sections fail to extinguish, going brown instead
Size: maximum 0·25 mm.

These data also clearly suggest arfvedsonite, except that in this case the optic axial plane is ∥(010), whilst it is normally ⊥(010) in both arfvedsonite and riebeckite. Sundius (1946) postulates a continuous series from arfvedsonite through kataphorite to eckermannite, as follows:

	(Fe,Mn):Mg		
Arfvedsonite ...	100:0 to 70:30	$\gamma = b$	$\alpha:c$ small, say $< 10°$
Kataphorite ...	70:30 to 30:70	$\gamma = b$ (variable)	$\alpha:c$ medium 15–36°
Eckermannite ...	30:70 to 0:100	$\beta = b$	$\alpha:c$ large 45–56°

Sundius also claims that kataphorite commonly has a normal symmetrical position of the optic axial plane with $\gamma = b$, but that this relationship is not always held. He postulates analogous variations in the refractive indices to those suggested above in accounting for the differences between types (i) and (ii) of intrusion A. Thus he suggests that the variation in the position of the optic axial plane is reasonable in view of the low birefringence, which would allow small modifications of the chemical composition to effect a change in the size of the chief indices. Bearing this in mind, and also the variation noted within the amphiboles of intrusion A, it is suggested that intrusion B contains a mineral of the arfvedsonite-kataphorite range, in which the optic axial plane is perpendicular to the usual direction.

Age of intrusions.

The field relations suggest that these arfvedsonite-rhyolites are dikes, but no direct evidence of age can be deduced, other than that they are younger than the Dalradian metamorphism. J. Phemister (1950, p. 368) has recently pointed out that in the case of the riebeckite-bearing dikes of Scotland '. . . no conclusion on the question of age can be reached on the basis of mineralogical similarity' with other Scottish rocks. It would seem to be even more dangerous to attempt such a correlation between sodic rocks in Donegal and Scotland.

Reference has been made to the fact that no analogous rocks have been discovered within the main suite of microgranitic dikes cutting the neighbouring Donegal granite. It has been noted in other areas that strongly sodic (particularly riebeckitic) rocks occur in close association with ring-complexes. Particular interest attaches to the present dikes because of the granitic ring-complexes of the Rosses (Pitcher, 1953), Ardara, and possible other foci on the west coast of Co. Donegal. Pitcher (1953, p. 155) describes the Rosses granite as being '. . . of the same general age as the Caledonian Newer Granites of Scotland'. It is

not impossible that these two phenomena are in fact related, and that the arfvedsonite-rhyolites represent a specialized magma intruded penecontemporaneously with the other varied microgranitic dikes. However, no definite proof of age exists.

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An occurrence of a regular mixed-layer clay-mineral.

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I. Introduction.

About twelve miles south-east of Burghersdorp in the Cape Province, South Africa, the unusual clay reported in this paper was first discovered on the farms 'Groenvlei' and 'Kuilfontein'. Later deposits were also uncovered on the farms 'Kapokkraal' and 'Kleinhoek'.

The clay-mineral appears to be of a very unusual type, namely, a regular mixed-layer clay-mineral (1:1 hydrous mica and montmorillonite). The mode of origin of the clay is also of interest in that it appears to be produced from a shale by hydrothermal alteration, due to intrusion of dolerite sheets.

The clay, owing to absence of iron, appears white in colour and burns white. It is thus being used in the ceramic industry as a member of an insulator body, where it acts as a bond in the green ware and as a flux upon firing of the product. Finely ground, the clay can also be used successfully as a filler for high-grade paper.

II. Occurrence.

Geologically, the area in which the clay occurs comprises the Upper Beaufort and the sandstones and shales of the Molteno beds with numerous intrusions of dolerite sheets. In all the known occurrences the clay appears at a contact zone between a thick dolerite sheet and the Molteno shales.

The white clay is usually found at or near the contact with a dolerite sheet. It is a compact material, with a hardness of 2, a smooth feel, and a conchoidal fracture. Black dendritic growths appear on some fracture surfaces. The thickness of this white material varies between 2 and 5 feet and then progresses into material with a cream or yellow colour. On moving farther away from the dolerite sheet the typical Molteno shales, yellow or grey in colour, are found.

Geologically, the appearance of the clay seems to be associated with

an intrusion of dolerite into the Molteno shale, and its probable genesis is a hydrothermal alteration of these shales. The hydrothermal alteration was presumably not very intense, but merely resulted in the progressive hydration of the illitic clay minerals of the Molteno shale, with accompanying removal of silica and iron. Without doubt more deposits will be uncovered in the future within the vicinity of the known occurrences which are extensive and already worked economically.

III. Chemical analysis.

In table I the chemical analyses are given of the white clays on the farms 'Kuilfontein' and 'Groenvlei' and also tabulated are analyses of the yellow clay and of Molteno shale from the deposit on the latter farm. Depths below the intrusion are indicated.

Table I. Chemical analyses of Burghersdorp clays.

	White clay.		Yellow clay.	Molteno shale.	
	Kuilfontein.	Groenvlei.	Groenvlei.	Groenvlei.	
	1 foot.	1 foot.	5 feet.	9 feet.	15 feet.
SiO_2	52·02	51·69	59·44	63·06	72·51
Al_2O_3	34·18	34·50	28·47	26·26	18·79
Fe_2O_3	0·58	0·19	1·36	1·22	0·59
TiO_2	0·68	0·68	0·76	0·70	0·84
MgO	0·41	0·16	0·17	—	0·10
CaO	1·44	1·56	1·15	0·94	0·71
Na_2O	1·02	1·03	1·15	0·49	0·17
K_2O	3·19	3·56	2·34	2·32	2·90
SO_3	0·23	0·12	0·06	0·27	0·03
gn. loss	6·79	5·96	5·47	5·50	3·66
	100·54	99·45	100·37	100·76	100·30
Base-exchange capacity	45 m.e.	48 m.e.	41·5 m.e.	24·8 m.e.	10·5 m.e.

(Analyses by D. Sampson on dry basis 110° C.)

The yellow clay differs essentially from the white clay in that its iron content is higher and that it contains free quartz (as proved by X-ray diffraction data). The underlying Molteno shale also shows a progressive increase in quartz content. It is clear that no sharp boundaries exist and that the Molteno shale at 9 feet is considerably altered. The sample at 15 feet may be regarded as unaltered shale.

As will be shown later, the only minerals which could be detected in the unaltered shale were quartz and illite. The relative proportions could not be measured, but, if it is assumed that free quartz forms about 45 % of the shale, the remainder of the analysis conforms reason-

ably well to that of an illite. If the various depths can be regarded as stages in the process of alteration, we can say that the main change, apart from the alteration of the clay-mineral, is removal of the large amount of free quartz. Iron is also removed in the white clay near the intrusion, but appears to accumulate at farther distances. Part of the potassium is also lost, since removal of free quartz would otherwise lead to the presence of 5 6 % K_2O in the altered material.

From the analysis of the white clay from 'Groenvlei' the following formula can be calculated:

$$\begin{matrix} \begin{pmatrix}\text{exchangeable}\\ 0{\cdot}016\text{Mg},\ 0{\cdot}07\text{Ca}\end{pmatrix} & \begin{pmatrix}\text{non-exchangeable}\\ 0{\cdot}04\text{Ca},\ 0{\cdot}13\text{Na},\ 0{\cdot}298\text{K}\end{pmatrix} \\ \uparrow & \\ [\text{Ti}_{0{\cdot}03},\text{Fe}_{0{\cdot}01},\text{Al}_{1{\cdot}95}]\ [\text{Al}_{0{\cdot}68},\text{Si}_{3{\cdot}32}]\ \text{O}_{10}[\text{OH}]_2 & \end{matrix}$$

On the evidence to follow, the mineral has a mixed-layer structure so that, if differences in chemical composition occur between the two types of layers, the above formula must be regarded as an average. The Mg^{++}, Ca^{++}, Na^{+}, and K^{+} are interlayer cations. Using the ammonium acetate Kjeldahl distillation method of base-exchange determination a base-exchange value of 48 m.e. per 100 g. clay dried at 110° C. was found. The ammonium-acetate leach liquor was also analysed and found to contain Mg^{++} and Ca^{++} in proportions corresponding to 46 m.e. per 100 g. clay, confirming the value from Kjeldahl distillation. This corresponds to all the Mg^{++} and about two-thirds of the Ca^{++} of the natural occurring clay. Analysis of the ammonium-acetate treated clay confirmed that this amount of Mg^{++} and Ca^{++} had been removed.

One-third of the Ca^{++} and all of the Na^{+} and K^{+} were found to be non-exchangeable by NH_4^{+} or any other cation. The 3·56 % of fixed potassium oxide is characteristic of illite, although distinctly smaller in percentage than in true illites. Non-exchangeable Na^{+} and Ca^{++} are presumably trapped between non-expanding layers along with the K^{+}.

The base-exchange capacity appears too high for an illite and too low for a montmorillonite, suggesting that the clay is in some way a mixture of the two. Many clays consist of mixtures (1) in such a way that discrete particles of the individual minerals occur, with no preferred geometric orientation of one clay particle with respect to its neighbours. Another type of mixing, however, first reported by Gruner (2), is the interstratification of parallel layers of separate minerals. In recent years such 'mixed-layer minerals' have frequently been reported (3, 4, 5), and it appears to represent a common mode of formation of clay minerals in deposits of hydrothermal origin (6, p. 325). Many earlier clay-

mineral species such as beidellite (7), bravaisite (4), &c., are now recognized as mixed-layer minerals.

Mixed-layer minerals are of two types: (i) those in which the succession of the two types of layers is random; and (ii) those in which two types of layers are arranged in a regular order. The distinction between these must be made on the grounds of X-ray diffraction data.

IV. X-ray diffraction data.

X-ray powder diffraction patterns were obtained with a Philips high-angle spectrometer goniometer, type 42201/2, with Co-$K\alpha$ radiation (λ 1·78890) and an automatically recording Brown Electronik strip-chart potentiometer.

From a total of 49 X-ray diffraction diagrams it was possible to determine the X-ray diffraction data for the pure clay-mineral (although a mixture in itself) as it occurs in the natural state (table II). The white clay usually contained no other admixed minerals except free quartz in the deposits on 'Kapokkraal'. In all the deposits, the underlying yellow-coloured clay and the Molteno shales respectively showed increasing amounts of free quartz.

Table II. X-ray data for Burgersdorp clay.

d.	Int.	*d.*	Int.	*d.*	Int.	*d.*	Int.
25·8*	vs	3·67	w	2·451	w	1·977	vw
12·4*	s	3·33	m	2·232	vw	1·683 / 1·646	w b
4·95	mw	3·09	mw	2·190	vw		
4·47	s	2·833	w	2·145	vw	1·492	m
4·30	mw	2·563	m	2·027	vw	1·287	w

* On ethylene glycol treatment the values increase to 28·1 and 13·6 Å. respectively.

The samples from the 'Groenvlei' occurrence, chemical analyses of which are given in table I, were used for comparison. Apart from peaks in the region of the basal spacing, all showed a diffraction pattern, which, except for the occurrence of quartz, corresponded completely to a dioctahedral illite or hydrous mica (8). The peaks representing the basal spacing and higher spacings are reproduced in fig. 1. The unaltered shale at 15 feet shows a well-defined peak at 9·99 A., and the clay mineral present must therefore be regarded as illite. In the sample at 9 feet, this peak is distinctly weaker and shows a broadening towards higher spacings. In the yellow clay, this process has advanced farther with the highest intensity of the broad peak at 12·4 Å. and the appearance of a new peak at a spacing of 25·8 Å. The white clay shows intense,

sharp peaks at 12·4 Å. and 25·8 Å. only. This indicates that the ma i
alteration in the clay-mineral according to the X-ray investigation is a
progressive hydration, resulting in an increase of the basal spacing.

Drying the clay at 145° C. and 500° C. showed a progressive moveme

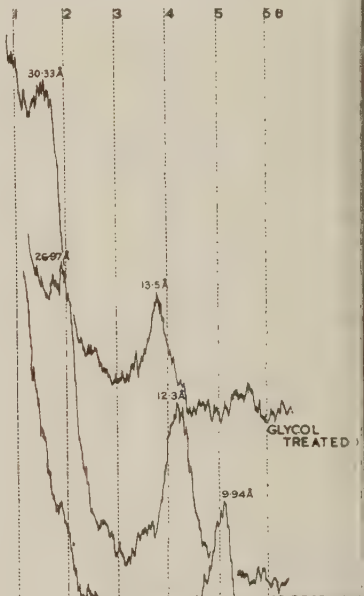

Fig. 1. Fig. 2.

Basal X-ray diffraction peaks of:

Fig. 1. The clay profile, showing progressive alteration of illite to a mixed-laye
clay-mineral.

Fig. 2. Mixed-layer clay-mineral after various treatments.

of the 12·4 A. X-ray diffraction peak to 10·47 Å. and 9·94 A. respec
tively, while the 25·8 Å. peak decreased in intensity and ultimatel
disappeared. The clay then slowly rehydrated to its original state whe
left at room conditions again. Ethylene glycol treatment caused a
increase of the 12·4 A. and 25·8 Å. values to 13·6 Å. and 28·1 Å. respec
tively. These properties again indicated the presence of a clay-minera
with an expandable lattice, i.e., of montmorillonite (fig. 2).

It would thus appear that X-ray data, in agreement with the chemica

nalysis and base-exchange properties, indicate the presence of both an litic or hydrous mica mineral and of a montmorillonitic mineral.

Attempts to obtain only a montmorillonite or illite typical basal X-ray diffraction peak value by $MgCl_2$ (9), $CaCl_2$ (4), or KOH (10) reatments of the original white clay failed.

If the two clay-minerals were present as a mixture, the basal reflection t 12·4 Å. should not appear. Instead, separate reflections at 10 Å. and 5·4 Å. should occur in the natural clay, and at 10 Å. and 17·7 Å. in the glycol-treated clay. It seems clear therefore that the present mineral is a mixed-layer type.

The mathematical theory of X-ray diffraction for random mixed-ayer structures has been presented by Hendricks and Teller (11), and further developed by Brown and MacEwan (8). In a random structure, an average basal spacing is to be expected, such as the 12·4 Å. and the 3·6 Å. (glycol-treated) in the present case. These spacings suggest that montmorillonite and hydrous mica layers are present in roughly a 1:1 ratio, and this is also in accord with the base-exchange capacity.

In a random stratification, no spacings larger than the average basal spacing are to be expected. If, however, the arrangement is regular, the diffraction effect is equivalent to a larger unit cell which is a multiple of the individual layers. In the present case, a distinct reflection at 25·8 Å. in the natural occurring white clay must be taken as proof of a regular structure, based on a 1:1 ratio. If this is regarded as 10–15·8 Å., the shift to 28·1 Å., i.e., 10–18·1 Å. on glycol treatment is in accord with expectations. So also is the disappearance of these reflections on heating, since a montmorillonite layer on dehydration presents a basal spacing at 10 Å. which does not differ significantly from that of an illite layer. The only previous record of a clay mineral with regular alternating layers is that of rectorite by Bradley (12), with a first-order spacing of 25 Å., shifting to 26·6 Å. with ethylene glycol. This was interpreted as a 1:1 ratio of pyrophyllite to vermiculite.

With a 1:1 ratio, the second-order reflection for the higher spacing falls in the same position as the average of the individual basal spacings and cannot therefore be distinguished from it. No evidence for a third-order reflection at 8·6 Å. was obtained with any sample.

If the regularity of the 1:1 arrangement were perfect the mean basal spacing should be exactly equal to half of the higher spacing, i.e., in this case it should be 12·9 Å. instead of 12·4 Å. The difference can only be accounted for if we postulate that the ratio of hydrous mica : montmorillonite is a little in excess of 1:1. The additional hydrous mica layers

would be inserted at random in the structure, and would give rise to a decrease of the mean basal spacing. Such insertions of course impair the regularity of the 1:1 arrangement, thereby tending to make the peak due to the higher spacing less sharp and less intense. Only a moderate excess of hydrous mica layers distributed in this way would suppress the higher spacing reflection altogether.

It is of interest to note that the greatest regularity was not found where alteration was most intense. The sample of white clay in fig. 1 was taken about 2 feet away from the intrusion, while a sample of white clay taken next to the intrusion is illustrated in fig. 2. It will be seen that for much the same second-order basal spacing, the higher spacing is distinctly larger but gives a much less well-defined peak. On glycol treatment it also shows a greater shift. It is not proposed to discuss the significance of these differences in detail, but it is clear that they correspond to a less regular structure.

When the Ca^{++}, Mg^{++}, H^{+}, NH_4^{+}, K^{+}, Na^{+}, Li^{+}, and Ba^{++} clays were prepared by treating the clay with either the acetate or chloride salts of these cations the basal reflections shifted to the extent of the amount of layers of water adsorbed (13) by the montmorillonite member of the mixed-layer mineral. The various exchangeable cations gave the following results: Ca^{++} 12·27, Mg^{++} 11·99, and H^{+} 12·51 Å., showing two molecular layers of water adsorbed by the expandable layer; while NH_4^{+} 10·8, K^{+} 10·8, Na^{+} 10·8, Li^{+} 11·0, and Ba^{++} 11·0 Å., indicated the presence of a monomolecular layer of water within the expandable layer.

V. Differential thermal analysis.

The automatic differential thermal analysis apparatus used in this laboratory has been described fully by Theron (14) and the differential thermal holder by the author (15) in an earlier publication. All the differential thermal curves were obtained by heating material dried at 80° C. at a rate of 15° C. per minute and using the same sensitivity scale (220 μv.) on the recording meter. From 38 differential thermal runs representative curves are reproduced in fig. 3, again of the profile at the deposit on 'Groenvlei'.

The Molteno shale at 15 feet gives a typical curve for illite, while the Molteno shale at 9 feet shows already some signs of alteration, namely, more intense endothermal peaks, and a slight change in shape of the second endothermal peak. The fully developed alteration is shown by the white clay with a curve that compares well with reported mixed-

yer differential thermal curves in the literature (6, 8). The yellow clay ave the same curve as the white clay and is not reproduced. The main ifferences between the curve for illite and that for the mixed-layer ineral is a general intensification of the reactions and the appearance f a new endothermic reaction at 700° C. in the latter mineral.

As in the case with the X-ay investigation, the differential hermal analysis results sub-antiate the theory of the lteration of the shale into the ixed-layer clay-mineral due to ydrothermal changes following ie dolerite intrusions.

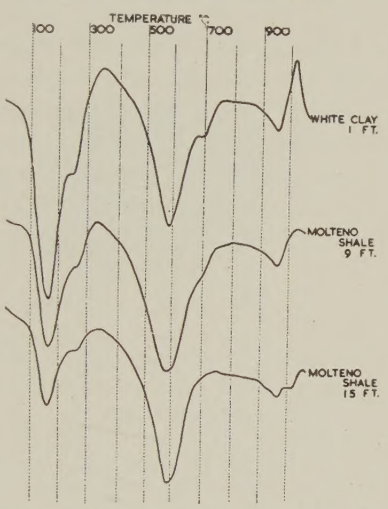

FIG. 3. Differential thermal curves of the clay profile, showing progressive alteration of illite to a mixed-layer clay-mineral.

DISCUSSION OF MODE OF ALTERATION.

From the facts in the previous ections it becomes possible to orm some picture of the mode f alteration of the original illite o a mixed-layer mineral. It eems reasonable to postulate hat this takes place by hydra-ion of alternate layers, accompanied by the removal of potassium rom such layers. In the shale at 9 feet the chemical analysis (table I) ndicates that the change has probably taken place to a marked extent. This is not necessarily in agreement with the X-ray diffraction data fig. 1), since replacement of the 10 Å. peak by a 12·4 Å. peak and the ppearance of a higher spacing are only to be expected when the altera-ion is nearly complete, i.e., when the hydrous mica:montmorillonite atio approaches unity throughout the whole of the clay-mineral. In he yellow clay, despite the marked changes in the basal X-ray dif-raction pattern, there is probably not much further alteration of the lay-mineral. The most complete change is evidenced by the white clay, ut even in this case, as already discussed, there is evidence that the tructure is not completely regular. This is to be expected from the node of formation, since this is bound to lead to faults in the regularity, .e., occasionally two hydrous mica layers will be sandwiched between nontmorillonite layers instead of a single layer.

It can well be understood that conditions next to the intrusion ar
too intense for alteration to take place in a completely regular manner
and this would explain the type of X-ray diagram shown in fig. 2.

According to the above view-point, every second layer of the origina
illite should remain undisturbed. In such a case, since sodium is non
exchangeable, the sodium to potassium ratio should not alter. Th
analyses in table I, however, indicate that there is a higher ratio o
sodium to potassium after alteration has taken place. If the sodium
content of 0·17 % is truly representative of the unaltered shale, it ha
to be admitted that this seems contrary to the theory suggested above

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